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A STATISTICAL THEORY ON THE TURBULENT DIFFUSION
OF GAUSSIAN PUFFS

T. Mikkelsen, S.E. Larsen and H.L. Pécseli

Abstract. The relative diffusion of a one-dimensional Gaussian cloud of particles is related to a two-particle covariance function $R_{abs}(\xi_{ij}, \tau) = \overline{u(x_i(t))u(x_i(t-\tau)-\xi_{ij})}$ in a homogenous and stationary field of turbulence. This two-particle covariance function expresses the velocity correlation between one particle (i) which at time t is in the position x_i , and another particle (j), which at the previous time $t-\tau$ is displaced the fixed distance ξ_{ij} relative to $x_i(t-\tau)$. For $\xi_{ij} = 0$, R_{abs} reduces to the Lagrangian covariance function of a single particle. Setting, on the other hand, the time lag τ equal to zero, a pure Eulerian fixed point covariance function results.

(Continue on next page)

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For diffusion times that are small compared to the integral time scale of the turbulence, simple expressions are derived for the growth of the clouds standard deviation $\sigma(t)$ by assuming that the wave number spectrum corresponding to the Eulerian space covariance $R_{abs}(\xi_{ij}, 0)$ can be expressed as a power law δk^p , where δ is a constant. For instance, by setting $p = -5/3$, an initially small cloud is found to growth as $\sigma^2(t) = (2\Gamma(\frac{2}{3})\delta)^{3/2} t^3$ in agreement with Batchelor's (1950) inertial subrange theory. Correspondingly, for the enstrophy cascade subrange in two-dimensional turbulence, for which case $p = -3$, the theory yields $\sigma^2(t) = \sigma_0^2 \exp(\delta t^2)$, where σ_0 denotes the initial size of the cloud.

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CONTENTS

	Page
1. INTRODUCTION	5
2. THEORY	13
2.1. Dispersion in a frame of reference attached to the centre of gravity	13
2.2. Kinematics of particle movements in a moving frame	20
3. TURBULENT DIFFUSION OF GAUSSIAN PUFFS	24
3.1. Relative diffusion equation	24
3.2. Spectral formulation of relative diffusion	45
3.3. Approximative solutions to the relative diffusion equation	49
3.4. Spreading of Gaussian puffs related to Eulerian power law spectra	53
4. APPLICATION TO ATMOSPHERIC DISPERSION	57
4.1. Relative diffusion within the inertial subrange ...	57
4.2. Relative diffusion within the enstrophy inertial subrange	60
4.3. Relative diffusion within the troposphere	63
5. DISCUSSION	68
6. CONCLUSION	75
REFERENCES	77
APPENDIX A: Important notation	80
APPENDIX B: Spectral definitions	86
FIGURE LEGEND	89

1. INTRODUCTION

The most important property of turbulent fluid motion is maybe its ability to disperse fluid particles which were initially close together. This is of practical importance for the dispersal and dilution of pollutants in the environment and is also of fundamental importance to the nature of turbulence.

The very first theories on eddy diffusion in the atmosphere put forward almost simultaneously by G.I. Taylor (1915) and L.F. Richardson (1922) were direct generalisations of the classical theory of molecular diffusion. They assumed that the mass effect of the eddies was entirely similar, except for a scale difference, to that of the molecules, thus it was suggested that an eddy-diffusivity of the order 10^{-2} to $10^7 \text{ m}^2 \text{ s}^{-1}$ should replace a molecular diffusivity of the order $10^{-5} \text{ m}^2 \text{ s}^{-1}$ in entirely similar differential equations. It became soon clear, however, that the difference between the eddy structure of a turbulent fluid and the molecular structure of a fluid at rest was more than one of scale. The failure of this early theory became evident by the enormous variations found in K , the eddy diffusivity. Richardson evaluated K for the diffusion of smoke over short distances, for the distribution of volcanic ash, and for the scatter of small balloons, and found K 's varying from 10^0 to $10^4 \text{ m}^2 \text{ s}^{-1}$. Other estimates varied from 10^{-2} to $10^7 \text{ m}^2 \text{ s}^{-1}$,

and in general it was found that K increased rapidly with the scale of the phenomenon. The need of an extended theory to express the observed differences led G.I. Taylor (1921) to formulate the problem of diffusion by continuous movement. In his contribution to the subject, G.I. Taylor extended the theory on the problem of the scatter caused by uncorrelated movements in a fluid to the case where a correlation exists between the motion of a particle at one instant and its motion at some subsequent time. By doing so, Taylor solved the problem of relating single particle dispersion in homogeneous turbulence to Lagrangian statistics of the velocity field.

The fundamentally different characteristics of two-particle statistics, or the statistics of a dispersing cloud of marked fluid in a turbulent field were first considered by F.L. Richardson (1926, 1929) and later by Batchelor (1950) and Brier (1950). Richardson (1926) pointed out that relative dispersion is an accelerating process in which an initially marked volume of fluid is spread at a rate depending upon its size. Richardson summarized various atmospheric diffusion data (over the range of 1 km to 10 km) and arrived at the "4/3-power law" for the relative, or instantaneous, diffusion coefficient K_R defined by

$$K_R = \alpha l^{4/3} \quad (1.1)$$

where l is the distance separating two typical marked fluid elements and α is a constant. A list of notation is contained in Appendix A.

To describe the shape characteristics of a dispersing cloud, F.L. Richardson (1926) introduced the distance neighbour function $q(l, t)$ defined by

$$q(l, t) = \frac{1}{A} \int_{-\infty}^{\infty} C(l+l', t) C(l', t) dl' \quad (1.2)$$

where

$$A = \int_{-\infty}^{\infty} C(l', t) dl'$$

and $C(l', t)$ is the instantaneous concentration distribution along a line l' at time t . The quantity $q(l, t)$ is an even function and its second and fourth moments are simply related to those of the concentration curve by

$$\sigma^2 = \frac{1}{2} \overline{l^2} \quad (1.3)$$

$$\mu = \frac{1}{2} \overline{l^4} - \frac{3}{4} (\overline{l^2})^2$$

where σ^2 and μ are the second and fourth moment of $C(l')$ about its centre of mass at a given time t .

Richardson also suggested the differential equation

$$\frac{\partial q}{\partial t} = \frac{\partial}{\partial l} \left(\alpha l^{4/3} \frac{\partial q}{\partial l} \right) \quad (1.4)$$

to describe the variable q . This has a solution (G.K. Batchelor, 1952)

$$q(l, t) = M \left(\frac{2}{3\pi^{1/2}} \right) \left(\frac{9}{4\alpha t} \right)^{3/2} \exp \left(- \frac{9l^{2/3}}{4\alpha t} \right), \quad (1.5)$$

$$l^2 = \frac{35}{9} \left(\frac{2}{3} \alpha t \right)^3$$

for the initial condition $q(l, 0) = M\delta(l)$ together with the constraint $\int_{-\infty}^{\infty} q(l, t) dl = M$, where $\delta(l)$ is the Dirac delta function. Note that the formulation by Richardson in Eq. (1.4) implies that the spreading of two marked fluid elements depends upon their instantaneous random separation l .

A theoretical interpretation of the empirical relation Eq. (1.1) was later given by Obukhov (1941) and Batchelor (1950, 1952) in terms of the universal similarity theory of Kolmogoroff. For the inertial subrange of high Reynolds number flow, Batchelor deduced that

$$K_R = c \epsilon^{1/3} l^{4/3} \quad (1.6)$$

where c is a constant of order unity and ϵ is the rate of energy dissipation.

The significance of introducing two-particle statistics in the relative dispersion problem was recognized by both Brier (1950) and Batchelor (1950) who independently demonstrated the involvement of the correlation between velocities of two different particles separated in both space and time. This two-particle Lagrangian correlation function is now well known to be fundamental

to the relative or cloud dispersion problem in the same way as single-particle Lagrangian correlation function is fundamental for the fixed frame diffusion problem. Following Batchelor (1950), the equation for the mean square separation of an arbitrary pair of particles is

$$\frac{d}{dt} \overline{l^2(t)} = 2 \int_0^t \overline{\{u_1(t) - u_2(t)\} \cdot \{u_1(\tau) - u_2(\tau)\}} d\tau. \quad (1.7)$$

where the subscripts identify the particles, u is the particles velocity component along the line l where also the spread $\overline{l^2}$ is measured, and overbars represent an ensemble average over a large number of realizations of the turbulent field and t and τ are two times.

Eq. (7) contains two types of velocity product. The first, of the form $u_1(t) \cdot u_1(\tau)$ refers to the same particle at two different times and thus represents a Lagrangian single particle velocity covariance. The second, of the form $u_1(t) \cdot u_2(\tau)$ involves one particle at time t and a second at time τ and is thus a two-particle Lagrangian covariance at different instants.

An alternative to P.L. Richardson's formula (Eq. (1.4)) to describe the shape characteristics of a dispersing cloud was also given by Batchelor (1952), in which the effective diffusivity depends upon the statistical quantity $\overline{l^2}$ rather than upon the random instantaneous separation l

$$\frac{\partial \bar{q}}{\partial t} = \alpha (\bar{l}^2)^{2/3} \frac{\partial^2 \bar{q}}{\partial \bar{l}^2} \quad (1.8)$$

The solution satisfying the same conditions as Eq. (1.5) is here

$$\bar{q}(\bar{l}, t) = \frac{1}{(2\pi \bar{l}^2)^{1/2}} \exp\left(-\frac{1}{2} \frac{\bar{l}^2}{\bar{l}^2}\right),$$

$$\bar{l}^2(t) = \left(\frac{2}{3} \alpha t\right)^3 \quad (1.9)$$

where the quantity \bar{q} denotes an ensemble-average value, taken over concentration distributions arising from the release of a large number of identical clouds of marked fluid.

The form of the two solutions, Eq. (1.5) and Eq. (1.9), are significantly different, this large difference allowed Sullivan (1971) to test the two hypotheses against each other, using relatively crude, but repeated observations of dye plumes. His results showed that the average of several instantaneous concentration distributions about their centre of mass of gravity were approximately Gaussian and the ensemble averaged distance-neighbour function to be of approximately Gaussian form. Thus the data were consistent with the theoretical description of Batchelor, Eq. (1.9).

Various attempts to experimentally verify Batchelor's (1950) theory on the two particle Lagrangian correlation function, Eq. (1.7) (Gifford 1957a,b; 1977), have so far not thrown light on the nature of this function, or its effect on relative disper-

sion. Only qualitative agreement is found with Batchelor's inertial range theory for small times

$$\overline{l^2} = \overline{l_0^2} + 2(\overline{u_1(0) - u_2(0)})^2 \cdot t^2 \quad (1.10)$$

and for intermediate times

$$\overline{l^2} = c \cdot \overline{u^2} t^3 \quad (1.11)$$

where l_0 is the initial separation of the pair of particles and c is a constant of order unity. However, various approximate forms of the two-particle Lagrangian correlation have been proposed (Brier 1950; Batchelor 1952; Smith and Hay 1961; C.J.P. Van Buijtenen 1982). Sawford (1982) compared the mean-square separation predictions from the first three of these and also from an approximation suggested by G.I. Taylor (see Batchelor 1952), in which the two-particle covariance for different instants is replaced by a simple product of a two-particle covariance at the same time and the single particle Lagrangian autocovariance function, R_L . That is,

$$\begin{aligned} \overline{u_1(t) \cdot u_2(\tau)} &= \overline{u_1(t) \cdot u_2(t) \cdot R_L(t-\tau)} \\ &= \overline{u_1(t) \cdot u_2(t)} \cdot \overline{u_1(\tau) \cdot u_1(t)} / \overline{u^2} \end{aligned} \quad (1.12)$$

By comparison with suitably documented observations, Sawford found this approximation to be the most appropriate.

In the chapter that follows the kinematics of particles involved in a relative diffusion process is discussed. In Chapter 3 follows then the derivation of a formula for the growth rate of a one-dimensional Gaussian puff (or cloud) of particles. In Chapter 4 is finally discussed the implications of the theory in Chapter 3 to various atmospheric dispersion problems.

Throughout the rest of this report it will be assumed that $t \geq \tau$ without loss of generality and the theory is restricted to scales large compared to the Kolmogorov scale $(\nu^3/\epsilon)^{1/4}$ (Batchelor 1950) so that the effects on molecular diffusion may be ignored.

2. THEORY

2.1. Dispersion in a frame of reference attached to the centre of gravity

Consider the release at time $t = 0$ of a cloud of marked fluid into a field of stationary and homogeneous turbulence. Let the observed concentration field at subsequent times of the experiment be given by $C(\underline{x}, t)$. This field is subject to the continuity equation, which in integral form reads

$$Q = \int C(\underline{x}, t) d\underline{x} \quad (2.1)$$

The quantity Q is the total amount of matter released with the puff. The volume integral extends over all space. The quantity $Q^{-1} \overline{C(\underline{x}, t)} d\underline{x}$ describes the probability of finding particles in the volume element $d\underline{x}$ surrounding the point \underline{x} , at time t . The first moment of the normalized concentration field $Q^{-1} C(\underline{x}, t)$ yields the instantaneous position $\underline{c}(t)$ of the centre of mass of the cloud

$$\underline{c}(t) = \frac{1}{Q} \int \underline{x} C(\underline{x}, t) d\underline{x} \quad (2.2)$$

Like any single "marked" fluid particle, $\underline{c}(t)$ executes random movements as a function of time in a turbulent environment. The velocity of the centre of mass position vector, $\underline{v}_{cm} = d\underline{c}/dt$,

follows from a differentiation of Eq. (2.2). By use of the continuity equation, this time in differential form

$$\frac{\partial C}{\partial t} = -\nabla \cdot (\underline{u} C(\underline{x}, t)) , \quad (2.3)$$

where \underline{u} is the velocity vector of the fluid and $\nabla \cdot$ is the divergence operator, use can be made of the fact that $\lim_{|\underline{x}| \rightarrow \infty} C(\underline{x}, t) = 0$, whereby Eq. (2.2) becomes

$$\underline{v}_{cm}(t) = \frac{1}{Q} \int \underline{u} C(\underline{x}, t) d\underline{x} \quad (2.4)$$

A relative coordinate system \underline{y} , attached to the puffs centre of mass \underline{c} , may now be defined by

$$\underline{y} = \underline{x} - \underline{c} \quad (2.5)$$

This "moving" frame of reference is exposed to continuous acceleration by the turbulence and is as such characterized as a non-inertial frame of reference.

The observed concentration field may as well be described in this "relative" frame of reference. Clearly, $C(\underline{y}, t) = C(\underline{x} - \underline{c}, t)$. The relative frame description $C(\underline{y}, t)$ differs only from the "fixed" frame description $C(\underline{x}, t)$ in the trivial point of a different coordinate origin. However, as will be shown, significant differences exist between the statistical properties of C as observed at a fixed \underline{x} and fixed \underline{y} , respectively.

The ensemble average of the velocity of the centre of mass vector \underline{V}_{cm} may be determined from Eq. (2.4).

$$\overline{\underline{V}_{cm}} = \frac{1}{Q} \int (\underline{\bar{u}} \overline{\underline{C} + \underline{u}' \underline{C}'}) d\underline{x} \quad (2.6)$$

Primes denote fluctuations, i.e. departures from the ensemble mean in an individual realization. The mean product $\overline{\underline{u}' \underline{C}'}$ is identified as a local turbulent flux vector. In a homogeneous field and provided that the cloud when released is symmetrical about the origin, this flux must be antisymmetrical, so that its space-integral is zero (Csanady, 1973, p. 86). Thus for symmetrically released clouds, for others at least approximately

$$\overline{\underline{V}_{cm}} = \frac{1}{Q} \int \underline{\bar{u}} \overline{\underline{C}}(\underline{x}, t) d\underline{x} \quad (2.7)$$

In the homogeneous field of consideration, the mean velocity $\overline{\underline{V}_{cm}}$ of the diffusing particles will be zero or constant. Without loss of generality, the 'fixed' coordinate \underline{x} can be allowed to drift with the mean velocity $\underline{\bar{u}}$, i.e. the coordinate \underline{x} can be chosen so as to make $\overline{\underline{V}_{cm}}(t) = \underline{0}$. By assuming this, the zeroth and first moments of the cloud, calculated on basis of the ensemble-average over many realizations of the flow, becomes in the fixed (\underline{x}) and the moving (\underline{y}) frames, respectively

$$Q = \int \overline{\underline{C}}(\underline{x}, t) d\underline{x} = \int \overline{\underline{C}}(\underline{y}, t) d\underline{y} \quad (2.8)$$

$$\underline{\bar{C}} = \int \underline{x} \overline{\underline{C}}(\underline{x}, t) d\underline{x} = \int \underline{y} \overline{\underline{C}}(\underline{y}, t) d\underline{y} = 0$$

Any physically meaningful difference between 'fixed' and 'moving' frame ensemble average concentration fields $\bar{C}(x,t)$ and $\bar{C}(y,t)$ are therefore confined to their second and higher moments.

The second moment of the concentration distribution in the x and y frames are also simply related. By use of the definition of the centre of gravity Eq. (2.2), we have for each of the three Cartesian coordinate components*

$$\begin{aligned}
 & \int_{-\infty}^{\infty} y^2 C(y,t) dy \\
 &= \int_{-\infty}^{\infty} (x-c)(x-c) C(x,t) dx \\
 &= \int_{-\infty}^{\infty} x^2 C(x,t) dx + c^2 \int_{-\infty}^{\infty} C(x,t) dx \\
 &\quad - 2c \int_{-\infty}^{\infty} x C(x,t) dx \\
 &= \int_{-\infty}^{\infty} x^2 C(x,t) dx - c^2 Q .
 \end{aligned} \tag{2.9}$$

*(Where all the variables refer to the same Cartesian coordinate component, specific designation of the individual components (1,2,3) have been omitted for simplicity).

On repeating a given release a large number of times, an ensemble average value of Eq. (2.9) may be obtained. When thus ensemble-averaged, the left hand side of eq. (2.9) may be identified as the componentwise, mean square spread of the cloud, calculated in the moving frame of reference, y

$$\overline{y^2}(t) = \frac{1}{Q} \int_{-\infty}^{\infty} y^2 \overline{C}(y,t) dy , \quad (2.10)$$

and the first term on the right hand side may be identified as the mean square spread of the particles in the 'fixed' frame of reference, x

$$\overline{x^2}(t) = \frac{1}{Q} \int_{-\infty}^{\infty} x^2 \overline{C}(x,t) dx \quad (2.11)$$

The last term, $\overline{c^2}(t)$ represents the mean square spread of the "centre of mass" movement of the puffs, also referred to the fixed frame of reference.

Eq. (2.9) can now be written as

$$\overline{x^2}(t) = \overline{y^2}(t) + \overline{c^2}(t) \quad (2.12)$$

which states that the spread, referred to an absolute frame of reference, of an ensemble of clouds which are released at $x = 0$, equals at time $t > 0$ the sum of the relative spread of the puff and the spread of the puffs centre of mass movement, referred to the absolute frame of reference. Clearly, $\overline{x^2}$ is always greater than either $\overline{y^2}$ or $\overline{c^2}$.

In the previous section it was mentioned that relative diffusion is closely related to the rate at which two arbitrary diffusing particles separate (cf. the discussion in connection with Eq. (1.7)). To establish a relationship between the mean square separation $\overline{\ell^2}$ of two diffusing particles and the mean square distance from the centre of gravity (Eq. (2.10)), let a point cloud be released at $t = 0$ at the origin of a fixed coordinate x , being parallel to the line ℓ , and consider the mean product

$$1/Q^2 \overline{C(x,t) C(x',t)} dx dx' \quad (2.13)$$

The quantity $Q^{-1} \overline{C(x,t)}$ is the probability that a marked fluid will be found at the distance x at successive times t . This is also equal to the probability of displacement x in time t for a single diffusing particle. The product may be regarded as the joint probability of finding marked fluid particles both at x and at x' , hence it is also equal to the joint probability of particle displacements for two diffusing particles x and x' , in time period t . Denoting the two-particle displacement probability density by $P(x,x',t)$, such that $P(x,x',t) dx' dx$ is the probability of finding one particle at x , and another at x' , we may also write

$$\overline{C(x,t) C(x',t)} = Q^2 P(x,x',t) \quad (2.14)$$

The second moment of $P(x,x',t)$ with respect to the separation vector $(x-x')$ yields the mean square separation $\overline{\ell^2}$ of two diffusing particles

$$\begin{aligned}
 \overline{l^2} &= \iint (x'-x)^2 P(x, x', t) dx' dx \\
 &= \frac{1}{Q^2} \iint (x'-x)^2 \overline{C(x, t) C(x', t)} dx' dx \\
 &= 2 \overline{x^2(t)} - 2 \overline{c^2(t)}
 \end{aligned} \tag{2.15}$$

By use of Eq. (2.12) this simply becomes

$$\overline{l^2} = 2 \overline{y^2(t)}, \tag{2.16}$$

which states that along an arbitrary coordinate direction, the mean square separation of two diffusing particles is just twice their mean square separation from the centre of mass.

The probability density $P(x, x', t)$ may also be regarded as specifying the probability of an absolute displacement x , and a relative displacement $\xi = x' - x$ of the two particles. Multiplying by Q and integrating over all displacement x remains the ensemble mean of the distance neighbour function mentioned in the previous paragraph

$$\begin{aligned}
 \overline{q}(\xi, t) &= Q \int P(x, x', t) dx \\
 &= \frac{1}{Q} \int \overline{C(x, t) C(x+\xi, t)} dx
 \end{aligned} \tag{2.17}$$

The integral over the concentration product can be determined for individual realizations and yields a somewhat smoothed picture of the distribution of particles within the cloud. As suggested by F.L. Richardson (1926), this ensemble average neighbour density constitutes a possible description of relative diffusion alternative to the mean concentration distribution in a moving frame (cf. Eq. (1.2)).

2.2. Kinematics of particle movements in a moving frame

Let the velocities of the marked fluid or suspended particles referred to the moving frame be $\underline{v}(v_1, v_2, v_3)$. From a differentiation of Eq. (2.5) it then follows that

$$\underline{v} = \underline{u} - \underline{V}_{cm} \quad (2.18)$$

From Eq. (2.8) we have $d\bar{c}/dt = \overline{\underline{V}_{cm}} = 0$ and, without loss of generality, we may assume that $\overline{\underline{u}} = 0$ (by measuring \underline{u} relative to a frame of reference moving with any mean motion of the ensemble).

Then, from Eq. (2.18) it is also clear that the ensemble-averaged velocity of a particle, relative to the centre of mass coordinate of the cloud is zero.

$$\overline{\underline{v}} = d\bar{\underline{y}}/dt = 0 \quad (2.19)$$

Because the relative velocity and displacement of the diffusing particles within the puff are related by the Lagrangian integral

$$\underline{y} = \int_0^t \underline{v}(t') dt' \quad (2.20)$$

an analogue Taylor's theorem, using relative velocities, can formally be derived. Along individual Cartesian coordinate directions, the mean square displacement of the cloud varies as

$$\frac{d\overline{\langle y^2 \rangle}}{dt} = 2\overline{\langle y \frac{dy}{dt} \rangle} = 2 \int_0^t \overline{\langle v(t)v(t') \rangle} dt' \quad (2.21)$$

where v is the component of the Lagrangian velocity vector \underline{v} that is parallel to y .

Two types of averaging are involved here. The overbars indicate as previously ensemble averaging over all realizations of the turbulent field whereas the brackets $\langle \rangle$ implies an average over all marked fluid or particles in the cloud. It must also be emphasized, however, that the relative velocity $v(t)$, in contrast to the absolute velocity $u(t)$ usually used with Taylor's theorem, does not constitute a stationary process. At the beginning when an initially small cloud is released, only the smallest turbulent eddies contribute to $v(t)$ and thereby to the growth, then increasingly larger ones, until the maximum eddy size is reached and exceeded. The velocity covariance $\overline{\langle v(t)v(t') \rangle}$ is thus not only a function of time lag $\tau = t-t'$, but depends also on the diffusion time t explicitly.

A modified Lagrangian correlation function can formally be introduced which is appropriate for the relative velocities of particles within the cluster, Csanady (1970)

$$r(\tau, t) = \frac{\overline{\langle v(t)v(t-\tau) \rangle}}{\langle v^2(t) \rangle} \quad (2.22)$$

The qualitative behaviour of this relative velocity correlation function is shown in Fig. 1.

At zero time lag $\tau = 0$, $r(\tau, t)$ has its maximum value of unity. As with the Lagrangian correlation functions of absolute velocities (in homogeneous and isotropic turbulence), $r(\tau, t)$ probably never becomes negative but remains a monotonically decreasing function of the time lag τ . Formally, $r(\tau, t)$ defines a Lagrangian integral time scale $t_r(t)$ appropriate for relative diffusion, which can be visualized as the shaded area in Fig. 1

$$t_r(t) = \int_0^t r(\tau, t) d\tau \quad (2.23)$$

The time of release of the cloud is here arbitrarily set equal to zero in the lower limit of the integral.

This relative Lagrangian time scale is characteristic for the average lifetime of eddies contributing to the movement of the particles relative to the centroid of the cloud. These eddies are ranging from a size comparable to the size of the cloud and down to the smallest length scale of the fluid, i.e. the

Kolmogorov scale $(\nu^3/\epsilon)^{1/4}$. Of these eddies, however, the ones of size comparable to the cloud will be the most energetic. This is true at least for diffusion in the range where the energy spectrum is a decreasing function of the wavenumber.

In this region the time scale $t_r(t)$ must be expected to be closely related to the decay time of eddies of size comparable to the size of the cloud. A simple estimate of $t_r(t)$ is $(\overline{y^2}/\overline{v^2})^{1/2}$. As the cloud grows, successively larger eddies begin to contribute, the larger the eddy, the longer is its "memory" or decay time. It is herefrom qualitatively understandable that t_r , and the mean square relative velocity $\overline{v^2}$ as well, must be increasing functions of the diffusion time t . Since $r(\tau, t)$ has the maximum value of unity and is a decreasing function for $\tau > 0$, an upper bound for the relative time scale is given by $t_r(\tau) \leq t$. Ultimately, when the cloud becomes so big that particles associated with it move independently of each other, t_r will cease to grow and becomes equal to the Lagrangian time scale of the fluid t_L . In this far field limit, also $\overline{v^2}$ will cease to grow and asymptotically approach the variance of the fluid, $\overline{u^2}$.

By combining Eq. (2.22) and Eq. (2.23), the second moment of the distribution function Eq. (2.21) may now be written as

$$\overline{y^2} = 2 \int_0^t \overline{v^2(t')} t_r(t') dt' \quad (2.24)$$

The equation represents a kinematic formulation of the relative mean square spread defined in Eq. (2.10).

3. TURBULENT DIFFUSION OF GAUSSIAN PUFFS

3.1. Relative diffusion equation

Here will be considered the dispersion of passive one-dimensional clouds or puffs, released from an instantaneous point source in a homogeneous and stationary field of turbulence. The particle density distribution function will, in accordance with common practice, be assumed to be Gaussian, and the growth of the cloud will be calculated in terms of the Gaussian standard deviation σ . By restricting the cloud dispersion to take place along a single, but arbitrarily oriented Cartesian component only, the analysis allows for calculating relative diffusion in situations where the turbulent field is not necessarily isotropic. This is of great practical importance. In the planetary boundary layer of the atmosphere, for instance, the turbulent field in the two horizontal component directions may, under certain conditions, be considered homogeneous and stationary but due to the presence of the ground, it is not isotropic on scales where relative diffusion of pollutants is of interest.

Chapter 2 led to a general kinematic formulation of the process of the relative diffusion of a cloud in the coordinate system moving with the centroid of the cloud. The starting point will here be the differential Equation (2.21) which applies as well to the calculation of the growth of a one-dimensional Gaussian

puff, the standard deviation of which is denoted by $(\overline{y^2})^{1/2} = \sigma(t)$

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \overline{v(t)v(t-\tau)} d\tau \quad (3.1)$$

As before, the particle velocity $v(t) = dy(t)/dt$ and the brackets implies an average over all the particles in the cloud, which here is assumed to have a Gaussian density distribution whereas the overbar indicates an ensemble averaging over the turbulent velocity field in question. The moving frame velocity covariance in Eq. (3.1) may next, by use of Eq. (2.18), be related to the fixed frame particle velocity u and the velocity of the centeroid V_{cm} .

$$\begin{aligned} \overline{v(t)v(t-\tau)} &= \overline{(u(t)-V_{cm}(t))(u(t-\tau)-V_{cm}(t-\tau))} \\ &= \overline{u(t)u(t-\tau)} - \overline{u(t)V_{cm}(t-\tau)} - \overline{u(t-\tau)V_{cm}(t)} \\ &\quad + \overline{V_{cm}(t)V_{cm}(t-\tau)} \end{aligned} \quad (3.2)$$

For convenience it is temporarily feasible to consider the Gaussian cloud as made up of a very large, but finite number N of individual particles. In this case the averaging over the particles in the cloud $\langle \rangle$ explicitly reads $1/N \sum_{i=1}^N$. The subsequent generalization back to the continuous particle distribution function is achieved by letting N approach infinitely. A reduction of the terms in Eq. (3.2) follows now from the fact that in homogeneous and stationary turbulence, Lagrangian auto-covariance functions of individual and simultaneously released

particles are identical. In the fixed frame, the i 'th particle's auto-covariance function reads $\overline{u_i(t)u_i(t-\tau)}$, where the suffix i refers to the i 'th particle of the cloud. In the moving frame, the same particle's auto-covariance function reads $\overline{v_i(t)v_i(t-\tau)}$.

The terms in Eq. (3.2) thereby becomes

$$\begin{aligned}\overline{v(t)v(t-\tau)} &= \frac{1}{N} \sum_{i=1}^N \overline{v_i(t)v_i(t-\tau)} = \overline{v(t)v(t-\tau)} \\ \overline{u(t)u(t-\tau)} &= \frac{1}{N} \sum_{i=1}^N \overline{u_i(t)u_i(t-\tau)} = \overline{u(t)u(t-\tau)} \\ \overline{u(t)v_{cm}(t-\tau)} &= v_{cm}(t-\tau) \frac{1}{N} \sum_{i=1}^N \overline{u_i(t)} = \overline{v_{cm}(t-\tau)v_{cm}(t)} \\ \overline{u(t-\tau)v_{cm}(t)} &= v_{cm}(t) \frac{1}{N} \sum_{i=1}^N \overline{u_i(t-\tau)} = \overline{v_{cm}(t)v_{cm}(t-\tau)}\end{aligned}\tag{3.3}$$

The first two of these equations states that the cloud-averaged ($\langle \rangle$) auto-covariance function, in moving and fixed coordinates respectively, equals the auto-covariance function of an individual particle. The quantity $1/N \sum_{i=1}^N u_i$ is analogue to the definition of the centre of mass velocity in Eq. (2.4).

By use of this, Eq. (3.2) now takes the simple form

$$\overline{v(t)v(t-\tau)} = \overline{u(t)u(t-\tau)} - \overline{v_{cm}(t)v_{cm}(t-\tau)}\tag{3.4}$$

Since the turbulence is assumed to be stationary, the Lagrangian auto-covariance $\overline{u(t)u(t-\tau)}$ must be independent of time t . This, however, is not the case for the relative velocity covariance, nor for the centre of mass velocity covariance function in Eq. (3.4).

Setting $\tau = 0$, Eq. (3.4) reduces to

$$\overline{u^2} = \overline{v^2(t)} + \overline{v_{cm}^2(t)}, \quad (3.5)$$

where the right hand side is explicitly written as functions of time in order to emphasize the non-stationarity of the terms. The equation states that the velocity variance of a particle or a fluid element, measured in the fixed frame of reference $\overline{u^2}$, is partitioned in a complementary manner between the variance of the velocity of the centre of mass of the cloud, and the variance of velocities relative to this, $\overline{v^2}$. The same result is easilier derived by ensemble averaging the square of Eq. (2.18) and making use of that $\overline{\langle v(t)v_{cm}(t) \rangle}$ is zero in the moving co-ordinate system.

An analogous Taylor's theorem, expressed in terms of relative coordinates and velocities was previously formulated in connection with Eq. (2.21). For diffusion referred to a fixed frame, this theorem applies to the spreading of the individual particles $\overline{dx^2}/dt$ (for which it originally was formulated) as well as to the position of the clouds centre of mass coordinate, $\overline{dc^2}/dt$. Therefore, the following set of equations applies to

the spreading along each of the three Cartesian coordinate directions

$$\begin{aligned} \frac{1}{2} \frac{\overline{dx^2}}{dt} &= \int_0^t R_{abs}(\tau) d\tau ; & R_{abs}(\tau) &\equiv \overline{u(t)u(t-\tau)} \\ \frac{1}{2} \frac{\overline{dc^2}}{dt} &= \int_0^t R_{cm}(t, \tau) d\tau ; & R_{cm}(t, \tau) &\equiv \overline{v_{cm}(t)v_{cm}(t-\tau)} \quad (3.6) \\ \frac{1}{2} \frac{\overline{dy^2}}{dt} &= \int_0^t R_{rel}(t, \tau) d\tau ; & R_{rel}(t, \tau) &\equiv \overline{v(t)v(t-\tau)} \end{aligned}$$

In Eq. (3.6), the Lagrangian covariance functions for the (absolute) velocity in the fixed frame x , for the velocity of the centre of mass coordinate c , and for the (relative) velocity in the moving frame y , have been abbreviated by $R_{abs}(\tau)^*$, $R_{cm}(t, \tau)$ and $R_{rel}(t, \tau)$, respectively.

By substituting the first of the Eqs. (3.3) into Eq. (3.1), and by subsequent use of Eq. (3.6), the following relation is easily obtained

*) To emphasize independence of the absolute time t , R_{abs} is defined here as a function of the time-lag τ , only.

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \frac{1}{2} \frac{d\overline{x^2}}{dt} - \frac{1}{2} \frac{d\overline{c^2}}{dt} \quad (3.7)$$

When integrated with respect to the time t , this equation becomes identical to the previous finding in Eq. (2.10).

In contrast to Eq. (2.10), however, the present equation constitutes a fundament on which the appropriate velocity covariance functions can be included to give the rate of growth of the cloud. A combination of Eq. (3.6) and Eq. (3.7) gives

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \{R_{abs}(\tau) - R_{cm}(t, \tau)\} d\tau \quad (3.8)$$

This equation, with $\sigma^2 = \frac{1}{2} \overline{l^2}$ (where $\overline{l^2}$ is the mean square separation of the particles) compares with the general formulation of the relative diffusion concept originally presented by Batchelor (1952) but also with Sawford (1982) (cf. Eq. (3) of the latter paper).

In Eq. (3.8), $R_{abs}(\tau)$ denotes the Lagrangian covariance function appropriate for single particle diffusion. In order to be able to integrate Eq. (3.8), however, also $R_{cm}(t, \tau)$ must be related to some fundamental statistical property of the turbulence. An attempt to do so is suggested in the following.

The centre of mass auto-covariance function is by definition given by

$$R_{cm}(t, \tau) = \overline{V_{cm}(t)V_{cm}(t-\tau)} = \overline{\langle u(t) \rangle \langle u(t-\tau) \rangle} \quad (3.9)$$

As previously discussed, the brackets in Eq. (3.9) symbolizes an (instantaneous) average over all the individual particles or marked fluid of the cloud. As shown above, this average can be performed by use of the instantaneous displacement distribution function of the cloud which, when referred to the fixed coordinate x , reads $Q^{-1} C(x, t)$. When multiplied by the (large) number N of particles that constitutes the cloud, $Q^{-1} C(x, t) dx$ denotes the (small) number of particles that occupies the position at time t between x and $x + dx$. At two fixed times, t and $t-\tau$, the velocity of the clouds centeroid as given by Eq. (2.4) therefore reads, respectively

$$\begin{aligned} V_{cm}(t) &= \frac{1}{Q} \int_{-\infty}^{\infty} u(x', t) C(x', t) dx' \\ V_{cm}(t-\tau) &= \frac{1}{Q} \int_{-\infty}^{\infty} u(x'', t-\tau) C(x'', t-\tau) dx'' \end{aligned} \quad (3.10)$$

and with these relations, the centre of mass covariance function in Eq. (3.9) becomes

$$\begin{aligned} R_{cm}(t, \tau) &= \\ \frac{1}{Q^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} &\overline{u(x', t) u(x'', t-\tau) C(x', t) C(x'', t-\tau)} dx' dx'' \end{aligned} \quad (3.11)$$

It has already been assumed that the form of the instantaneous displacement distribution function of the cloud $Q^{-1} C(x,t)$ develops in a self-similar way as function of time. In accordance with general practice, this distribution was taken to be Gaussian and thereby normal-distributed around the centeroid $c(t)$ of the cloud, with a standard deviation $\sigma(t)$

$$Q^{-1} C(x,t) = \frac{1}{\sqrt{2\pi} \sigma(t)} \exp\left\{-\frac{1}{2} (x-c(t))^2 / \sigma^2(t)\right\} \quad (3.12)$$

Upon inserting this in Eq. (3.11), however, the averaging over the turbulent field represented by the overbar still have to extend over the displacement distribution functions because the centeroid $c(t)$ moves around in a random manner as a function of time. But by use of the substitution $x = c+y$, the frame of reference can be changed from fixed (x) to moving (y) coordinates. In the moving frame, the Gaussian particle density distribution function, $G_{\sigma(t)}(y,t)$ becomes

$$G_{\sigma(t)}(y,t) = Q^{-1} C(c+y,t) = \frac{1}{\sqrt{2\pi} \sigma(t)} \exp\left\{-\frac{1}{2} y^2 / \sigma^2(t)\right\} \quad (3.13)$$

In addition hereto the following relation

$$Q^{-1} C(x,t) dx = G_{\sigma(t)}(y,t) dy \quad (3.14)$$

simply states that the number of particles in a small line element is not influenced by changing the frame of reference from fixed to moving coordinates.

The velocity of the centroid corresponding to Eq. (3.10) now becomes, with the moving coordinate y as independent variable

$$V_{CM}(t) = \int_{-\infty}^{\infty} u(y'+c, t) G_{\sigma}(t)(y', t) dy' \quad (3.15)$$

$$V_{CM}(t-\tau) = \int_{-\infty}^{\infty} u(y''+c, t-\tau) G_{\sigma}(t-\tau)(y'', t-\tau) dy''$$

and analogous to Eq. (3.11), the centre of mass covariance function now becomes

$$R_{CM}(t, \tau) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} u(y'+c, t) u(y''+c, t-\tau) G_{\sigma}(t)(y', t) G_{\sigma}(t-\tau)(y'', t-\tau) dy' dy'' \quad (3.16)$$

As a consequence of the change of frame of reference the stochastic variable c is removed from the distribution functions and the averaging over the turbulent field therefore now only affects the velocity covariance $u(y'+c, t)u(y''+c, t-\tau)$, as Eq. (3.16) shows. This covariance function will now be the subject to further investigation. It expresses the time-averaged (Eulerian) correlation of fluid velocity, measured at the two fixed points y' and y'' in the moving coordinate system at the two times t and $t-\tau$, respectively. The situation is shown in Fig. 2.

With the purpose of relating this fixed point velocity covariance to some more fundamental property of the turbulent flow, however, the underlying Lagrangian diffusion process of the problem has to be investigated.

In Fig. 3 is shown the Lagrangian trajectory $y_i(t)$ of a particle or marked fluid (i) that at the previous time $t-\tau$ was in the position $y_i(t-\tau)$. In the moving frame of reference, the displacement $\Delta y_i = y_i(t) - y_i(t-\tau)$ constitutes a stochastic process, having a distribution function $G_{\Delta y_i}$ as shown. The quantity $\overline{\Delta y_i^2}$ equals the i'th particle contributions to the cloud spreads in the period of time between $t-\tau$ and t . The growth of the cloud in the time interval between $t-\tau$ and t is therefore the collective result of the motion of all the particles motion over that period of time. Taking the distribution function for the individual particles, $G_{\Delta y_i}$, as identical and independent Gaussians will now be shown to be consistent with the Gaussian distribution function $G_\sigma(t)$ assumed for the particle density of the cloud.

That the distribution function $G_{\Delta y_i}$ is independent of any neighbour particles implies that $\overline{\Delta y_i \Delta y_j} = 0$ for $i \neq j$. Therefore, the distribution function $G_\sigma(t)$ can be calculated as a superposition of the spread from the individual particles. With the continuous distribution functions in question, this superposition leads to the integral (Mikkelsen et al. (1982))

$$G_\sigma(t)(y) = \int_{-\infty}^{\infty} G_{\Delta y_i}(Y-Y_0) G_\sigma(t-\tau)(Y_0) dY_0 \quad (3.17)$$

With $G_\sigma(t)$ and $G_\sigma(t-\tau)$ inserted as Gaussian distributions having standard deviations equal to $\sigma(t)$ and $\sigma(t-\tau)$, respectively, $G_{\Delta y_i}$ can easily be solved by a Fourier transform of the integral equation (3.17) to be another Gaussian having a standard deviation squared given by

$$\overline{\Delta y_i^2} = \sigma^2(t) - \sigma^2(t-\tau) \quad (3.18)$$

Instead of a priori assuming that the instantaneous cloud density distribution $G_\sigma(t)$ is Gaussian, it could alternatively have been assumed that the individual particles displacement distribution function in the moving frame, $G_{\Delta y_i}$, are identical and independent Gaussians, with a standard deviation as given by Eq. (3.18). From Eq. (3.17) it then follows that an initial Gaussian-distributed cloud, with standard deviation $\sigma(t-\tau)$, would evolve Gaussian at all subsequent times with standard deviation $\sigma(t)$. It can be claimed that the Gaussianness of the relative displacement process Δy_i , together with the relation

$$\overline{\Delta y_i \Delta y_j} = \begin{cases} \sigma^2(t) - \sigma^2(t-\tau) & \text{for } i = j \\ 0 & \text{for } i \neq j \end{cases} \quad (3.19)$$

is the fundamental assumption of the present theory, and that a Gaussian cloud results as a consequence hereof.

The requirement that any two particles disperse uncorrelated in the moving frame (i.e. $\overline{\Delta y_i \Delta y_j} = 0$ for $i \neq j$, no matter how close they are, appears to be rather restrictive in a realistic turbulent field. Where the cloud consists of a very large number of particles, however, in which case a continuum description of the turbulence applies, the requirement corresponding to $\overline{\Delta y_i \Delta y_j} = 0$ for $i \neq j$ is

$$l_T(t) \ll \sigma(t) \quad (3.20)$$

Here, l_r is the relative integral length scale of the turbulence, which in terms of the relative velocity v can be defined as

$$l_r(t) = \overline{v^2(t)}^{-1} \int_0^{\infty} \overline{v(y,t)v(y+\xi,t)} d\xi \quad (3.21)$$

Even though the inequality $l_r \ll \sigma$ imposes strong limitations on the turbulent field, it is not as restrictive as the corresponding two-particle requirement, especially not when the cloud becomes large. With this picture of the relative diffusion process in mind, it is now possible to continue the investigation of the velocity covariance $\overline{u(y',t)u(y'',t-\tau)}$ in Eq. (3.16).

In close analogy to the turbulent spreading of contaminant particles, also the turbulent field itself can be considered as consisting of a very large, but numerable number M of small fluid elements or fluid particles.

Suppose that the i 'th of these fluid particles is in the position $y_i = y'$ at the time t . The i 'th particle Lagrangian velocity $u(y_i(t))$ will then equal the Eulerian velocity $u(y',t)$ in this point and at that time. Equivalently, if the j 'th fluid particle at time $t-\tau$ is in the position $y_j = y''$, its Lagrangian velocity equals the Eulerian velocity of that point, $u(y_j(t-\tau)) = u(y'',t-\tau)$.

Now consider the situation in Fig. 4 which shows the trajectories of a pair of fluid particles (i) and (j), the separation of which at time $t-\tau$ is given by ξ_{ij} , both in the fixed (x) and in the moving (y) frame as well

$$\begin{aligned}\xi_{ij}(t-\tau) &= x_i(t-\tau) - x_j(t-\tau) \\ &= y_i(t-\tau) + c(t-\tau) - (y_j(t-\tau) + c(t-\tau)) \\ &= y_i(t-\tau) - y_j(t-\tau)\end{aligned}\tag{3.22}$$

Suppose one knew the conditional joint probability distribution $S(y_i(t) = y' | y_i(t-\tau) = y_j(t-\tau) + \xi_{ij}, y_j(t-\tau) = y'')$ for finding the fluid particle (i) in the position y' at time t and the fluid particle (j) in the position y'' at time $t-\tau$, with the condition that the separation of the two particles, at time $t-\tau$, is given by the fixed distance $\xi_{ij}(t-\tau) = y_i(t-\tau) - y_j(t-\tau)$.

The contribution from this particle pair (i) and (j) to the total covariance $\overline{u(y',t)u(y'',t-\tau)}$ could then be calculated as

$$\begin{aligned}S(y_i(t) = y' | y_i(t-\tau) = y_j(t-\tau) + \xi_{ij}(t-\tau), y_j(t-\tau) = y'') \\ \times \overline{u(y_i(t) | y_i(t-\tau) = y_j(t-\tau) + \xi_{ij}(t-\tau))u(y_j(t-\tau))}\end{aligned}$$

where the ensemble-averaged covariance function of the two particles velocity

$$\begin{aligned} \overline{u(y_i(t)|y_i(t-\tau) = y_j(t-\tau) + \xi_{ij}(t-\tau))u(y_j(t-\tau))} \\ \equiv \overline{u(y_i(t))u(y_i(t-\tau) - \xi_{ij}(t-\tau))} \end{aligned} \quad (3.23)$$

also is subject to the condition that the particle pair separation, at time $t-\tau$, equals the fixed distance ξ_{ij} .

The moving frame fixed point covariance function $\overline{u(y',t)u(y'',t-\tau)}$ can then in principle be obtained as the sum of pair contributions from all possible values of the separation ξ_{ij} of the fluid. This leads to the summation over all values of (i) and (j):

$$\begin{aligned} \overline{u(y',t)u(y'',t-\tau)} = \\ \sum_i^M \sum_j^M S(y_i(t)=y' | y_i(t-\tau) = y_j(t-\tau) + \xi_{ij}(t-\tau), y_j(t-\tau)=y'') \\ \times \overline{u(y_i(t))u(y_i(t-\tau) - \xi_{ij}(t-\tau))} \end{aligned} \quad (3.24)$$

In this equation, the velocity u of a fluid particle is not influenced by a change of reference from moving to fixed coordinates. In the fixed coordinate (x), the two particle covariance function in Eq. (3.23) and Eq. (3.24) therefore also reads

$$\overline{u(y_i(t))u(y_i(t-\tau) - \xi_{ij}(t-\tau))} = \overline{u(x_i(t))u(x_i(t-\tau) - \xi_{ij}(t-\tau))} \quad (3.25)$$

with the condition that $x_i(t-\tau) = x_j(t-\tau) + \xi_{ij}(t-\tau)$. As Fig. 5a shows, Eq. (3.25) expresses the correlation between the velocity of a fluid particle (i) at time t in the arbitrary posi-

tion x_i , and the velocity at time $t-\tau$ of the fluid particle (j) that is displaced the distance $\xi_{ij}(t-\tau)$ relative to $x_i(t-\tau)$.

Alternatively, by referring the fixed distance ξ_{ij} separating the two particles to time t as shown in Fig. 5b, rather than to time $t-\tau$, the covariance function between the two particles alternatively reads $\overline{u(x_j(t-\tau))u(x_j(t)+\xi_{ij}(t))}$, where now $x_i = x_j(t)+\xi_{ij}(t)$. In the stationary and homogeneous turbulent field of consideration, these two alternative definitions must be identical, since the situation in Fig. 5b follows immediately from a time reversal of the situation in Fig. 5a. Moreover, these covariance functions will be independent of both the fluid particles absolute position x , as well as of the absolute time t . This leaves a function of the time lag τ and the separation ξ_{ij} only, which will be denoted as

$$\begin{aligned} R_{abs}(\xi_{ij}, \tau) &= \overline{u(x_i(t))u(x_i(t-\tau)-\xi_{ij}(t-\tau))} \\ &= \overline{u(x_j(t-\tau))u(x_j(t)+\xi_{ij}(t))} \end{aligned} \quad (3.26)$$

Setting $\xi_{ij} = 0$ reduces this two-particle covariance to the Lagrangian auto-covariance function of a single particle: $R_{abs}(0, \tau) = R_{abs}(\tau)$, where $R_{abs}(\tau)$ was defined in Eq. (3.6). On the other hand, by setting $\tau = 0$, a pure Eulerian space-covariance results, for which the fixed separation distance ξ_{ij} is along the same direction as the velocity component u .

With both ξ_{ij} and τ set equal to zero, the two-particle covariance function yields the total energy $\overline{u^2}$ of the turbulence.

$R_{abs}(\xi_{ij}, \tau)$ defines, with $\xi_{ij} = 0$, a fixed frame Lagrangian integral time scale of the turbulence through

$$t_L = (\overline{u^2})^{-1} \int_0^\infty R_{abs}(0, \tau) d\tau \quad (3.27)$$

Also, a (fixed frame) Eulerian integral length scale for the turbulence can be defined through

$$L_E = (\overline{u^2})^{-1} \int_0^\infty R_{abs}(\xi_{ij}, 0) d\xi_{ij} \quad (3.28)$$

The two-particle covariance function $R_{abs}(\xi_{ij}, \tau)$ resembles somehow the two-particle Lagrangian covariance $\overline{u_1(t)u_2(\tau)}$ discussed introductionally in connection with Eq. (1.7). But where this covariance is conditioned for two fluid particles, which at the time of release is located at the source position, the covariance in Eq. (3.26) is conditional with respect to a fixed particle separation ξ_{ij} but at an arbitrary time, $t-\tau$.

It remains to investigate the joint probability distribution $S(y_i(t) = y' | y_i(t-\tau) = y_j(t-\tau) + \xi_{ij}(t-\tau), y_j(t-\tau) = y'')$ in Eq. (3.24) for finding the i 'th fluid particle at y' at time t , and the j 'th fluid particle at y'' at time $t-\tau$, with the condition that $y_i(t-\tau) = y_j(t-\tau) + \xi_{ij}(t-\tau)$. We can do so with the assumption about the behind-laying diffusion process, namely that the individual fluid particles in the relative frame follow identical and independent Gaussian statistics. On this basis, the probability that the i 'th of a total of M fluid particles will be in the position y' , with the condition that it at the previous

time $t-\tau$ was in the position $y_i(t-\tau) = y_j(t-\tau) + \xi_{ij}(t-\tau)$, is simply given by $1/M G_{\Delta y_i}(y' - y_i(t-\tau))$, with $G_{\Delta y_i}$ given by Eq. (3.17) and Eq. (3.18). Independent hereof, the probability for finding the j 'th fluid particle in the position y'' at time $t-\tau$ simply is $1/M$.

For the turbulent field considered, the following relations therefore applies

$$\begin{aligned}
 S(y_i(t) = y' | y_i(t-\tau) = y_j(t-\tau) + \xi_{ij}, y_j(t-\tau) = y'') \\
 &= 1/M^2 G_{\Delta y_i}(y' - y_i(t-\tau)) \\
 &= 1/M^2 G_{\Delta y_i}(y' - (y_j(t-\tau) + \xi_{ij})) \\
 &= 1/M^2 G_{\Delta y_i}(y' - y'' - \xi_{ij}) \quad (3.29)
 \end{aligned}$$

By substituting this, together with the two-particle covariance function in Eq. (3.26), the following expression is obtained for the fixed point velocity covariance in Eq. (3.24)

$$\overline{u(y', t) u(y'', t-\tau)} = \sum_{i=1}^M \sum_{j=1}^M 1/M^2 G_{\Delta y_i}(y' - y'' - \xi_{ij}) R_{abs}(\xi_{ij}, \tau) \quad (3.30)$$

If the linear extension of a fluid particle is denoted by d , and if the number of fluid particles between $x_i(t-\tau)$ and

$x_j(t-\tau)$ is denoted by $n = i-j$, the particle pair separation can be written as $\xi_{ij} = dn$.

Further, it is legal to calculate the double sum in Eq. (3.30) as a sum over all possible values of i and j where the difference $n = i-j$ is fixed, followed by a sum over all n , viz.

$$\overline{u(y', t)u(y'', t-\tau)} = \sum_{n=-M}^M \sum_{i=n+j}^M 1/M^2 G_{\Delta y_i}(y'-y''-dn) R_{abs}(dn, \tau) \quad (3.31)$$

Both $G_{\Delta y_i}$ and R_{abs} are decreasing functions of their argument dn . Therefore, by going to the limit for very large M , corresponding to an extension of the turbulent field to infinity on both sides of the diffusing cloud, only the differences for which $n \ll M$ will contribute to the double sum in Eq. (3.31). With n fixed at a value much smaller than M , the sum over $i = n+j$ approximately equals M times the argument in Eq. (3.31) and only the sum over the differences n remains

$$\overline{u(y', t)u(y'', t-\tau)} = \sum_{n=-M}^M 1/M G_{\Delta y_i}(y'-y''-dn) R_{abs}(dn, \tau) \quad (3.32)$$

Finally, by considering the extension of the individual fluid particles small relative to the Kolmogorov scale of the turbulence, the pair separation $\xi_{ij} = \Delta n$ can be considered a continuous independent variable ξ , and in its equivalent integral form, Eq. (3.32) becomes

$$\overline{u(y', t) u(y'', t - \tau)} = \int_{-\infty}^{\infty} G_{\Delta y_i}(y' - y'' - \xi) R_{abs}(\xi, \tau) d\xi \quad (3.33)$$

With this result it is now possible to calculate the centre of mass covariance function in Eq. (3.16). With the standard deviation of $G_{\Delta y}$ as given in Eq. (3.18), the following integral has to be evaluated

$$R_{cm}(t, \tau) = \iiint_{-\infty}^{\infty} R_{abs}(\xi, \tau) G_{\Delta y_i}(y' - y'' - \xi) G_{\sigma}(t)(y', t) G_{\sigma}(t - \tau)(y'', t - \tau) dy' dy'' d\xi \quad (3.34)$$

By keeping ξ fixed, the remaining two integrals is simply a (double) convolution of two Gaussian distribution functions. The result hereof is another Gaussian with standard deviation equal to the square root of the sum of the individual variances: $\{(\sigma^2(t) - \sigma^2(t - \tau)) + \sigma^2(t) + \sigma^2(t - \tau)\}^{1/2} = \sqrt{2} \sigma(t)$. Thereby, the final expression for the centre of mass covariance function becomes

$$R_{cm}(t, \tau) =$$

$$\int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \frac{1}{2\sqrt{\pi} \sigma(t)} \exp \left\{ -\frac{1}{4} \frac{\xi^2}{\sigma^2(t)} \right\} d\xi \quad (3.35)$$

When an initially small puff is released, σ is much smaller than l_E whereby $R_{cm}(t, \tau) \approx R_{abs}(0, \tau)$. This implies that the centre of mass covariance function, and thereby the centre of mass spread, equals that of a single particle in this limit.

In the other limit, when σ has grown to a size much bigger than the length scale l_E , $R_{cm}(t, \tau)$ becomes small compared to $R_{abs}(\tau)$. This implies that the centre of mass dispersion $\overline{c^2}$ becomes negligible in this far field limit, and that the relative diffusion (σ^2) is entirely dominated by single particle diffusion ($\overline{x^2}$).

When the centre of mass covariance function Eq. (3.35) is inserted in Eq. (3.8), an implicit formula for the growth of a Gaussian puff results

$$\begin{aligned} \frac{1}{2} \frac{d\sigma^2}{dt} &= \int_0^t \left\{ R_{abs}(0, \tau) \right. \\ &\quad \left. - \int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \frac{1}{2\sqrt{\pi} \sigma(t)} \exp \left(-\frac{1}{4} \frac{\xi^2}{\sigma^2(t)} \right) d\xi \right\} d\tau \end{aligned} \quad (3.36)$$

In the previous chapter (Eq. (2.24)), the cloud spread was expressed in terms of a mean square relative velocity $\overline{v^2(t)}$ and a relative Lagrangian time scale $t_r(t)$, as

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \overline{v^2(t)} \cdot t_r(t) \quad (3.37)$$

From Eq. (3.35) with $\tau = 0$, and from Eq. (3.4), the mean square relative velocity can now be identified as

$$\overline{v^2(t)} = R_{abs}(0,0) - \int_{-\infty}^{\infty} R_{abs}(\xi,0) \frac{1}{2\sqrt{\pi} \sigma(t)} \exp\left(-\frac{1}{4} \frac{\xi^2}{\sigma^2(t)}\right) d\xi \quad (3.38)$$

Equivalently, the relative correlation function $r(t,\tau)$ defined in Eq. (2.22), explicitly becomes

$$r(t,\tau) = \left(\overline{v^2(t)}\right)^{-1} \left\{ R_{abs}(0,\tau) - \int_{-\infty}^{\infty} R_{abs}(\xi,\tau) \frac{1}{2\sqrt{\pi} \sigma(t)} \exp\left(-\frac{1}{4} \frac{\xi^2}{\sigma^2(t)}\right) d\xi \right\} \quad (3.39)$$

With this correlation function given, the relative time scale $t_r(t)$ is easily obtained by an integration of $r(t,\tau)$ with respect to τ , as defined in Eq. (2.23).

3.2. Spectral formulation of relative diffusion

It is possible to introduce a spectral representation of the two-particle covariance function $R_{abs}(\xi, \tau)$ defined in Eq. (3.26). The spectrum $S(k, \omega)$, where k is wavenumber and ω is frequency is defined through the Fourier transform

$$S(k, \omega) = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \exp(-i(k\xi + \omega\tau)) d\xi d\tau \quad (3.40)$$

In Appendix B is shown that the single-particle Lagrangian spectrum $S_L(\omega)$, which is obtained by setting $\xi = 0$, is related to $S(k, \omega)$ through

$$\overline{u^2} S_L(\omega) = \int_{-\infty}^{\infty} S(k, \omega) dk \quad (3.41)$$

and also that the (fixed point) Eulerian spectrum $S_E(k)$, which results by setting $\tau = 0$, is related through

$$\overline{u^2} S_E(k) = \int_{-\infty}^{\infty} S(k, \omega) d\omega \quad (3.42)$$

The inverse Fourier transform corresponding to Eq. (3.40) is defined as

$$R_{abs}(\xi, \tau) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i(k\xi + \omega\tau)) dk d\omega, \quad (3.43)$$

It is herefrom seen that the single-particle Lagrangian covariance function $R_{abs}(0, \tau)$ can be represented as

$$\begin{aligned} R_{abs}(0, \tau) &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i\omega\tau) dk d\omega \\ &= \int_{-\infty}^{\infty} S_L(\omega) \exp(i\omega\tau) d\omega \end{aligned} \quad (3.44)$$

With these definitions, the growth rate of the cloud in Eq. (3.36) now becomes

$$\begin{aligned} \frac{1}{2} \frac{d\sigma^2}{dt} &= \int_0^t \left\{ \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i\omega\tau) d\omega dk \right. \\ &\quad - \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i(k\xi + \omega\tau)) \frac{1}{2\sqrt{\pi} \sigma(t)} \\ &\quad \times \exp\left(-\frac{1}{4} \frac{\xi^2}{\sigma^2(t)}\right) d\omega dk d\xi \Big\} d\tau \end{aligned} \quad (3.45)$$

The integration over ξ of the second term on the right hand side is an inverse Fourier transform of the Gaussian distribution, i.e.

$$\begin{aligned} \int_{-\infty}^{\infty} \frac{1}{2\sqrt{\pi} \sigma(t)} \exp\left(-\frac{1}{4} \frac{\xi^2}{\sigma^2(t)}\right) \exp(ik\xi) d\xi \\ = \exp\left(-k^2 \sigma^2(t)\right) \end{aligned} \quad (3.46)$$

By use of this in Eq. (3.45), the following equation results for growth of a Gaussian cloud, expressed in terms of the spectrum $S(k, \omega)$ of the turbulence

$$\begin{aligned} \frac{1}{2} \frac{d\sigma^2}{dt} = & \int_0^t \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \left(1 - \exp(-k^2 \sigma^2(t)) \right) \\ & \times \exp(i\omega\tau) d\omega dk d\tau \end{aligned} \quad (3.47)$$

By specifying $S(k, \omega)$, this equation can, numerically at least, be solved for $d\sigma/dt$ as a function of time t , and thereby also, upon a further integration over time from zero to t , for the cloud size $\sigma(t)$.

In Eq. (3.36) and in Eq. (3.45) as well, the term in the parenthesis $\{ \}$ equals the relative velocity covariance from Eq. (3.6) $\overline{v(t)v(t-\tau)}$. Analogous to the procedure used to arrive at Eq. (3.38), it is found by setting $\tau = 0$ that the mean square relative velocity, in the spectral representation, can be expressed as

$$\begin{aligned} \overline{v^2(t)} = & \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(\omega, k) \left(1 - \exp(-k^2 \sigma^2(t)) \right) d\omega dk \\ & = \int_{-\infty}^{\infty} \overline{u^2} S_E(k) \left(1 - \exp(-k^2 \sigma^2(t)) \right) dk \end{aligned} \quad (3.48)$$

This shows the important result that the mean square relative velocity of the expanding cloud is entirely related to the Eulerian space spectrum $S_E(k)$.

The relative correlation function, Eq. (3.39) correspondingly becomes, in terms of the spectrum $S(k, \omega)$

$$r(t, \tau) =$$

$$\overline{v^2(t)}^{-1} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(\omega, k) \left(1 - \exp(-k^2 \sigma^2(t))\right) \exp(i\omega\tau) d\omega dk \quad (3.49)$$

The relative time scale $t_r(t)$ is as before obtainable from an integration with respect to τ , as defined in Eq. (2.23).

The equation for growth, Eq. (3.47), will next be considered in the limit where the cloud size σ is large compared to the length scale l of the turbulence. Then, for all relevant values of k , the quantity $1 - \exp(-\sigma^2 k^2) \approx 1$ and by use of Eq. (3.41), there results in this limit

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \int_{-\infty}^{\infty} S_L(\omega) \exp(i\omega\tau) d\omega d\tau \quad (3.50)$$

Integrating twice with respect to time yields

$$\sigma^2(t) = t^2 \int_{-\infty}^{\infty} S_L(\omega) \frac{\sin^2(\frac{\omega t}{2})}{(\frac{\omega t}{2})^2} d\omega \quad (3.51)$$

This is simply G.I. Taylor's formula for single particle diffusion. It is seen, not surprisingly, that the different behaviour of the spread of a cloud, when compared with that of a single particle, is closely related to the spatial correlation of the turbulence.

In the limit where also the time t is large compared to the time scale t_L , Eq. (3.51) reduces to the usual far field limit $\sigma^2 = 2\overline{u^2} t_L t$, appropriate for single particle dispersion.

3.3. Approximative solutions to the relative diffusion equation

Here will first be investigated the implications of an approximation similar to that suggested by G.I. Taylor, see Eq. (1.12). Suppose that the two-particle covariance function in Eq. (3.26) can be replaced by a simple product of a fixed point Eulerian correlation function at time t : $\rho_E(\xi) = \overline{u(x,t)u(x+\xi,t)}/u^2$ and a single-particle Lagrangian auto-correlation function $\rho_L(\tau) = \overline{u(x_i(t))u(x_i(t-\tau))}/u^2$, in which case

$$R_{abs}(\xi, \tau) = \overline{u^2} \rho_E(\xi) \rho_L(\tau) \quad (3.52)$$

Even though Sawford (1982) found this type of approximation to be the best appropriate in his comparison, this approximation cannot in general be valid, and it is unlikely that it is particularly good except perhaps when τ is small compared to τ_L . The Fourier transform in Eq. (3.20) consequently gives, with this approximation,

$$S(k, \omega) = \overline{u^2} S_E(k) S_L(\omega) \quad (3.53)$$

where

$$S_E(k) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \rho_E(\xi) \exp(-ik\xi) d\xi \quad (3.54)$$

and

$$S_L(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \rho_L(\tau) \exp(-i\omega\tau) d\tau \quad (3.55)$$

When Eq. (3.53) is substituted into Eq. (3.47) an subsequent integration over ω results in

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \rho_L(\tau) \left\{ \overline{u^2} \int_{-\infty}^{\infty} S_E(k) [1 - \exp(-k^2 \sigma^2(t))] dk \right\} d\tau \quad (3.56)$$

However, as before, the term in the parenthesis $\{ \}$ equals the mean square relative velocity $\overline{v^2(t)}$, cf. Eq. (3.48). The remaining integral over τ is, when comparison is made with Eq. (3.37), identified as the relative Lagrangian time scale $t_r(t)$.

Consequently, based on the approximation in Eq. (3.52), the following set of equations for the growth of a Gaussian cloud results

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \overline{v^2(t)} \cdot t_r(t) \quad (3.57)$$

where

$$\overline{v^2(t)} = \overline{u^2} \int_{-\infty}^{\infty} S_E(k) [1 - \exp(-k^2 \sigma^2)] dk \quad (3.58)$$

and

$$t_r(t) = \int_0^t \rho_L(\tau) d\tau \quad (3.59)$$

A consequence of the "factorization" of $R_{abg}(\xi, \tau)$ into Eulerian and Lagrangian correlation functions is that the relative time scale becomes identical to the time scale appropriate for single-particle diffusion. The mean square relative velocity, however,

is here, as well as under more general conditions (Eq. (3.48)), found to be related exclusively to the Eulerian properties of the turbulence.

One question that remains to be investigated is to what extent the estimate of the relative time scale in Eq. (3.59) applies to common turbulence.

Starting with the limit for large times where $t \gg t_L$, the relative time scale $t_r(t)$ in Eq. (3.59) becomes equal to t_L as it properly should, when the particles move independently of each other. In the small time limit, on the other hand, the approximate solution to Eq. (3.59) yields

$$t_r(t) = t \quad \text{for} \quad t \ll t_L \quad (3.60)$$

since $\rho_L(\tau) \approx 1$ for small time lags. It can be examined to what extent this limiting value is consistent with the more general solution, Eq. (3.47), viz.

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \left(1 - \exp(-k^2 \sigma^2(t)) \right) \exp(i\omega\tau) d\omega dk d\tau \quad (3.61)$$

An integration over the time lag τ here gives

$$\frac{1}{2} \frac{d\sigma^2}{dt} = t \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \frac{\sin(\omega t)}{\omega t} \left(1 - \exp(-k^2 \sigma^2(t)) \right) d\omega dk \quad (3.62)$$

But when the time t is sufficiently small, the sinc function $\sin(\omega t)/\omega t$ remains close to unity for all values of the angular frequency ω , where $S(k, \omega)$ contributes to the integral, see Fig. 3. Therefore, applicable in the small time limit the following approximation must apply

$$\int_{-\infty}^{\infty} S(k, \omega) \frac{\sin(\omega t)}{\omega t} d\omega \approx \int_{-\infty}^{\infty} S(k, \omega) d\omega = \overline{u^2} S_E(k) \quad (3.63)$$

With this approximation, Eq. (3.62) becomes in the limit for $t \ll t_L$

$$\begin{aligned} \frac{1}{2} \frac{d\sigma^2}{dt} &= t \overline{u^2} \int_{-\infty}^{\infty} S_E(k) [1 - \exp(-k^2 \sigma^2(t))] dk \\ &= t \cdot \overline{v^2}(t) \end{aligned} \quad (3.64)$$

It is seen that also the small limit value for $t_r(t)$ from Eq. (3.60) is consistent with the general solution in Eq. (3.47).

For values of t in the interval between the near and the far field limits, the degree of approximation associated with $t_r(t)$ when estimated from Eq. (3.59), depends on the statistical dependence between the two variables ω and k . If ω and k are totally independent of each other, then is $S(\omega, k) = \overline{u^2} S_E(k) S_L(\omega)$, and consequently is the correlation $\overline{\omega k} = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \omega k S(\omega, k) d\omega dk$ equal to zero. On the other hand, zero correlation between ω and k in a particular turbulent field is only a necessary, and not a sufficient condition for independency, and thereby also for the applicability of Eq. (3.59).

There are situations, however, where it is not necessary to be concerned about the general applicability of Eq. (3.59). This is when the cloud growth is dominated entirely by the Eulerian properties of the turbulence, which is the case when the function $\overline{v^2(t)}$ plays an all dominant role for the spread in a relatively short period of time after the release at $t = 0$. In that case $t_r(r) \approx t$ ($\ll t_L$) is a reasonably good approximation for the relative time scale and the growth of the cloud can be calculated simply on the basis of Eq. (3.64).

3.4. Spreading of Gaussian puffs related to Eulerian power law spectra

When considering diffusion times that are small relative to the Lagrangian integral time scale of the turbulence t_L , it was shown above that the growth of the Gaussian puff is determined by the simple set of equations

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \overline{v^2(t)} \cdot t_r(t) \quad (3.65)$$

$$\overline{v^2(t)} = \overline{u^2} \int_{-\infty}^{\infty} S_E(k) \left(1 - \exp(-k^2 \sigma^2(t)) \right) dk$$

$$t_r(t) = t, \quad \text{for } t \ll t_L.$$

In the limit for small times, only the Eulerian properties of the turbulence (through the wavenumber spectrum $S_E(k)$) therefore comes into play. The set of equations (3.65) will now be investigated analytically by assuming that the Eulerian wavenumber

spectrum is given as a power law $S_E(k) = \delta k^p$, where δ is a constant of dimension $m^{(1+p)}$. Spectra characterized by $p \geq -1$ results in divergence of the otherwise normalized integral $\int_{-\infty}^{\infty} S(k)dk = 1$. Such powers can consequently be included in the analysis only as subranges of limited extension. A power law representation of the Eulerian wavenumber spectrum $S_E(k)$ is also of relevance only over limited ranges of wavenumbers. For instance, at very small wavenumbers ($k \sim 0$), the theoretical spectrum tends to be flat ($p = 0$), and approaches asymptotically the amplitude level $\sim \ell/\pi$.

For cases where the power p is within the interval: $-3 < p < -1$, the second of the set of equations (3.65) can be integrated by parts to give

$$\overline{v^2}(t) = - \frac{2\overline{u^2}\delta}{(p+1)} \Gamma\left(\frac{p+3}{2}\right) \sigma^{-(p+1)}, \quad \text{for } -3 < p < -1$$

(3.66)

where Γ denotes the gamma function.

For cases where $p \leq -3$, an analytical solution to the equation for $\overline{v^2}$ does not seem possible. Therefore an approximation of the high pass filter $(1 - \exp(-\frac{1}{2} k^2 \sigma^2(t)))$ by Heaviside's step function, has been introduced*

*This corresponds to a "top hat" rather than a Gaussian cloud.

$$H(\sigma) = \begin{cases} 0 & \text{for } k < 1/\sigma \\ 1 & \text{for } k \geq 1/\sigma \end{cases} \quad (3.67)$$

Hereafter, the relative velocity variance simply becomes

$$\overline{v^2}(t) = - \frac{2\overline{u^2}\delta}{(p+1)} \sigma^{-(p+1)}, \quad \text{for } p \leq -3 \quad (3.68)$$

The differential equation for $\sigma(t)$ in Eq. (3.65) is now readily solved.

The following basically different solutions are found, all of which are applicable only in the limit $t \ll t_L$.

i) for $-3 < p < -1$

$$\sigma(t) = \{ct^2 + \sigma_0^{1/q}\}q$$

ii) for $p = -3$

$$\sigma(t) = \sigma_0 \exp\left(\frac{1}{2} \overline{\delta u^2} t^2\right) \quad (3.69)$$

iii) for $p < -3$

$$\sigma(t) = \{\mathfrak{C} t^2 + \sigma_0^{1/q}\}q$$

Here, $q = 1/(3+p)$, $\mathfrak{C} = -\overline{u^2}\delta(3+p)/(1+p)$ and $c = \mathfrak{C} \Gamma((p+3)/2)$.

σ_0 is the initial size of the cloud, i.e. $\sigma(t = 0)$. In order that the solution iii) for $p < -3$ applies, it must in addition be required that $t < t_{\max}$, where $t_{\max} = (\sigma_0^{1/q}/|\mathfrak{C}|)^{1/2}$. The

limit $t = t_{\max}$, however, is never reached with finite size clouds.

The behaviour of $\sigma(t)$ in the phase of spread, where the initial puff size σ_0 is an important parameter, can also be deduced from Eq. (3.65) by substituting Eqs. (3.66) for $\overline{v^2}(0)$ with $\sigma = \sigma_0$. For $t \ll \{\sigma_0^2/\overline{v^2}(0)\}^{1/2}$, a second-order expansion of the initial spread reads

$$\sigma^2(t) = \sigma_0^2 + \overline{v^2}(0) \cdot t^2 \quad (3.70)$$

In form this equation is similar to Eq. (1.10), and is thus in accordance with the result of Batchelor's similarity theorem in the near field limit.

Within the time interval described by Batchelor as "intermediate", i.e. when viscosity and the initial puff size are no longer of dominating importance, but before the integral time scale t_L becomes an important scaling parameter, the first of the Equations (3.69) yields

$$\sigma(t) = c_9 t^{2/(3+p)} \quad (3.71)$$

where the constraints are: $-3 < p < -1$ and $\{\sigma_0^2/\overline{v^2}(0)\}^{1/2} \ll t_L$.

In the following chapter the implications to atmospheric dispersion of the set of Equations (3.65) and their solutions Eq. (3.69) will be discussed.

4. APPLICATION TO ATMOSPHERIC DISPERSION

4.1. Relative diffusion within the inertial subrange

Turbulence in the inertial subrange of the atmospheric boundary layer is often represented in terms of Eulerian wavenumber spectra in the non-normalized form

$$\overline{u^2} S_E(k) = \alpha \epsilon^{2/3} k^{-5/3} \quad (4.1)$$

Here, α is a constant of order unity and ϵ being the rate of dissipation of energy. Setting $\delta = \alpha \epsilon^{2/3} / \overline{u^2}$ and $p = -5/3$, Eq. (3.71) for the growth of a cloud becomes

$$\sigma^2(t) = (2 \Gamma(\frac{2}{3}) \alpha)^{3/2} \epsilon t^3 \quad (4.2)$$

applicable for "intermediate" times only as defined in Eq. (3.71). When compared with Eq. (1.11), this result is also found to be in agreement with Batchelor's inertial subrange theory on relative diffusion.

For the case of homogeneous and isotropic turbulence, Tennekes and Lumley (1972) suggest the value of the constant $\alpha = \frac{9}{55} \times 1.5 = 0.246$ for the wavenumber spectrum $S_E(k)$ in question. (It should be emphasized that the proper one-dimensional spectrum to be used here is the so-called longitudinal spectrum, and not the corresponding transverse spectrum, see Tennekes and Lumley (1972))

p. 251 for precise definitions). This is because the velocity u is parallel to the particle separation ξ in Eq. (3.26).

For inertial subrange isotropic and homogeneous turbulence, the prediction for the spread of a Gaussian puff therefore becomes

$$\begin{aligned}\sigma^2(t) &= (1.34 \times 2 \times \frac{9}{55} \times 1.5)^{3/2} \epsilon t^3 \\ &= .534 \frac{\overline{u^2}}{t_L} t^3\end{aligned}\tag{4.3}$$

where the dissipation rate ϵ for later comparison has been replaced by $\overline{u^2}/t_L$.

Independently, F.B. Smith (1968) and F. Gifford (1981) have derived corresponding formulas for the instantaneous spread of a plume at small times $t \ll t_L$

$$\sigma^2 = \frac{2}{3} \epsilon t^3\tag{4.4}$$

Their numerical coefficient is slightly larger than the coefficient found in Eq. (4.3). Their models, however, describe the spread of individually released particles, the velocity of which in the fixed frame is governed by a Langevin equation with a specified initial velocity, common to all the particles released. Their model result (Eq. 4.4) thus describes the ensemble averaged spread of conditionally released single particle diffusion rather than a real two-particle or relative diffusion process. Further, their model result is a consequence of an assumed Lagrangian exponential correlation function, the Fourier transform of which, when expressed in Eulerian terms, becomes

$$\overline{u^2} S_E(k) = \frac{\overline{u^2}}{\pi} \frac{l}{1+(lk)^2} \quad (4.5)$$

This spectrum is representative for a $k^{-5/3}$ law only in a rather limited wavenumber interval in the neighbourhood of $(kl)^2 = 5$. In this case, Eq. (4.5) can be approximated by

$$\overline{u^2} S_E(k) = \frac{5^{5/6}}{6\pi} \overline{u^2} l^{-2/3} k^{-5/3} \quad (4.6)$$

Setting $\alpha = 5^{5/6}/6\pi = 0.203$ the here derived relative diffusion model, Eq. (4.3), yields a result which compares with an exponential correlation function

$$\sigma^2 = 0.401 \epsilon t^3 \quad (4.7)$$

In this case also, a notably smaller coefficient is found compared to the conditional single-particle result of Eq. (4.4).

When two particles simultaneously are deployed from a source with negligible (but non-zero) initial separation, both of them are immersed into one and the same coherent eddy structure. Their motion will thus remain to be coherent over a longer period of time than will be the case with single released particles, immersed into individual eddy structures and correlated through a common initial velocity only. Being more correlated, the two simultaneously released particles will not diffuse as rapid as the independently released particles. This constitutes a possible explanation for the somewhat different c efficiencies found in Eqs. (4.4) and (4.7).

4.2. Relative diffusion within the enstrophy inertial surange

Two-dimensional turbulence theory has attracted wide-spread interest among meteorologists following the work of Kraichnan (1967) and others. The theoretical studies of Kraichnan of two-dimensional turbulence have shown that a source of energy and enstrophy (half-squared vorticity) isolated at wavenumber k_i leads to a wavenumber spectrum with a discontinuity at k_i . For $k < k_i$ energy is cascaded to lower wavenumbers and $S_E(k) \propto \epsilon^{2/3} k^{-5/3}$ and for $k > k_i$, enstrophy is cascaded to larger wavenumbers and $S_E(k) \propto \eta^{2/3} k^{-3}$, where η is the enstrophy cascade rate. In the latter range, the characteristic time scale T_c is $\eta^{-1/3}$. In contrast to eddy time scales in three-dimensional turbulence, this two-dimensional time scale, characteristic for the small eddies in two-dimensional flow, is independent of the scale of motion. In the atmosphere, T_c is typically ~ 1 day. Several authors have provided evidence for the existence of the k^{-3} law in large scale atmospheric spectra down to scales ~ 100 km (see, for instance, K.S. Gage (1979) for a recent summary).

By dimensional analysis, J.T. Lin (1972) obtained an exponential power law for relative diffusion in the enstrophy cascade range, by postulating that the relative diffusivity depends on the local mean square relative distance $\overline{l^2}$ and the enstrophy cascade rate η . By dimensional analysis

$$\frac{1}{2} \frac{d\overline{l^2}}{dt} = \gamma \eta^{1/3} \overline{l^2} \quad (4.8)$$

$$\overline{l^2} = l_0^2 \exp(2t/T_C) , \quad \text{for } t \gg T_C$$

where γ is an order of unity dimensional coefficient and l_0^2 the initial separation of two diffusing particles. Since, in Lin's dimensional analysis, T_C is considered a relevant time scale, it is implicitly assumed that $t \gg T_C$ in Eq. (4.8).

On the other hand, in the limit where the diffusion time t is small relative to T_C , t itself must be a proper scaling parameter for the relative time scale, i.e. $t_r \propto t$ (as also can be seen from Eq. (2.23)). The mean square relative velocity $\overline{v^2}$ scales then with the mean square separation and the fixed time T_C , so the bigger the separation, the bigger is also the relative variance, $\overline{v^2}$. Based upon the relative diffusivity $\overline{v^2} \cdot t_r$, dimensional analysis now gives

$$\frac{1}{2} \frac{d\overline{l^2}}{dt} = \gamma \frac{\overline{l^2}}{T_C^2} \cdot t \quad (4.9)$$

$$\overline{l^2} = l_0^2 \exp(t^2/T_C^2) , \quad \text{for } t \ll T_C$$

In the more familiar case of single-particle diffusion, characterized by an integral time scale t_L and a constant variance $\overline{u^2}$, dimensional analysis also yields two basically different solutions for the spread $\overline{x^2}$, analogous to Eqs. (4.8) and (4.9). When

$t \gg t_L$, the rate of growth $1/2 \overline{dx^2}/dt$ is proportional to the (absolute) diffusivity $u^2 \cdot t_L$ whereas, when $t \ll t_L$, it is proportional to $u^2 \cdot t$. This gives rise to the two well-known sub-ranges for the spread of a single particle: $\overline{x^2} \propto t$ and $\overline{x^2} \propto t^2$, respectively.

By comparing the solution Eq. (3.69) for $p = -3$ with the dimensional analysis, Eq. (4.9) it is found that the two solutions are consistent in that they have identical forms and that both of them applies to times that are small relative to the time scale of the turbulence.

In order to be able to compare the here suggested turbulent diffusion model with the dimensional result, Eq. (4.8), the time scale $t_T(t)$ in Eq. (3.65) is now set equal to T_C corresponding to the limit where $t \gg T_C$. Integrating the first of the equations (3.65) with $\overline{v^2}(t)$ as given by Eq. (3.68) for $p = -3$ results in

$$\sigma(t) = \sigma_0 \exp(t/T_C) \quad \text{for } t \gg T_C \quad (4.10)$$

This is consistent with the result of J.T. Lin's (1972) dimensional analysis for relative diffusion in the enstrophy cascade subrange. Eq. (4.10) applies to situations where the diffusion time t is large compared to the turbulent time scale T_C . At the same time, the puff size, σ , must be small compared to the length scale l of the turbulence.

Figure 7 shows a summary of the four different regimes of diffusion predicted with a k^{-3} power law. The spectrum is assumed to be constant ($\propto k^0$) for $k < 1/l$. Note that the asymptotical values of $\sigma(t)$ equals that of single-particle diffusion, when $\sigma > l$. This example emphasizes the importance of distinguishing between length and time scales, when dealing with relative diffusion.

4.3. Relative diffusion within the troposphere

A schematic one-dimensional wavenumber spectrum has in Fig. 8 been composed from the literature, showing the different subranges previously discussed. The diffusion of an initially small Gaussian cloud starts in the 3-dimensional isotropic inertial subrange and grows from here into the reverse energy cascading $k^{-5/3}$ inertial range of two-dimensional turbulence. By associating a sink rather than a source for enstrophy and energy at the 1000-km scale shown, the empirical data composed in Fig. 8 becomes consistent with the theory of Kraichnan (1967) previously discussed. After reaching a size $\sigma \sim 10^6$ m, the cloud growth into the k^{-3} enstrophy cascade subrange and ultimately, on the 10^7 m scale, the spectrum is assumed to level off.

By choosing the mean small scale energy dissipation rate as small as $1-2 \cdot 10^{-4} \text{ m}^2 \text{ s}^{-3}$ the spectrum becomes almost a straight line over the interface between two- and three-dimensional turbulence. This occurs because the universal constant for the two-dimensional upscale transport spectrum, α_{II} is much larger than

for the three-dimensional decay spectrum α_I . For the one-dimensional, longitudinal spectrum shown is chosen: $\alpha_I = 0.25$, $\alpha_{II} = 2.2$ and $\epsilon = 0.8 \cdot 10^{-3} \text{ m}^2 \text{ s}^{-3}$. At $k = 2\pi/1000 \text{ m}^{-1}$, $\overline{u^2 S_E(k)} = 10 \text{ m}^3 \text{ s}^{-2}$ and the rate of energy injection at $\tau \sim 1000 \text{ m}$ is $\overline{u^2} dS_E/dt = 3.1 \cdot 10^{-5} \text{ m}^2 \text{ s}^{-3}$. With the sink at $\tau = 10^6 \text{ m}$, also the time scale $T_C = \eta^{-1/3}$ can be determined to be of the order ~ 17 hours. The energy of the spectrum in the $-5/3$, the -3 and the flat part is $3\pi \text{ m}^2 \text{ s}^{-2}$, $99\pi \text{ m}^2 \text{ s}^{-2}$ and $200\pi \text{ m}^2 \text{ s}^{-2}$, respectively and the corresponding length scale $S(o)/\pi \approx 340 \text{ km}$.

By calculating $\sigma(t)$ in the $-5/3$ subranges on basis of Eq. (4.3), the clouds travel time t will exceed the enstrophy integral time scale T_C already at the $\sim 10^5 \text{ m}$ scale. Eq. (4.10), applicable for $t \gg T_C$, is hence the appropriate formula, rather than Eq. (4.9), for determination of the asymptotical form of $\sigma(t)$ in the -3 enstrophy cascade subrange.

Based on the formulas, Eq. (3.57)-(3.59), and on the spectrum in Fig. 8, not only the asymptotical form, but also the inter-regional growth of the Gaussian cloud can be determined on basis of the spectrum in Fig. 8. However, the Eulerian wavenumber spectrum does not give information on the Lagrangian correlation coefficient $\rho_L(\tau)$ and therefore neither on the relative time scale t_r in Eq. (3.59). For this reason, the following simple model for t_r is proposed for use here

$$t_r(t) = \frac{t_L}{1+t_L/t} \quad (4.10)$$

The quantity t_L is here the parameter value of the integral time scale appropriate for large scale dispersion (Gifford, 1982). The suggested function has the appropriate asymptotical forms, i.e. $t_r \approx t$ for $t \ll t_L$ and $t_r = t_L$ for $t \gg t_L$ as discussed previously in connection with Eq. (3.59) and Eq. (3.60). The condition $t_r \leq t$ is also fulfilled by Eq. (4.10). In addition, as long as $t_r(t)$ is chosen as a smooth and monotonically increasing function of time, its specific form influences only the growth marginally.

By use of the following set of substitutions

$$\delta^2 = \sigma^2 / \alpha^2 \quad \text{where} \quad \alpha^2 = \overline{u^2} t_L^2 \quad (4.11)$$

$$\tilde{t} = t / t_L ; \quad \tilde{t}_r = t_r / t_L = \tilde{t} / (\tilde{t} + 1) ,$$

Eq. (3.59) can now be written in the following non-dimensional form, appropriate for numerical integration

$$\frac{1}{2} \frac{d\tilde{\sigma}^2}{d\tilde{t}} = \tilde{t}_r(\tilde{t}) \int_{-\infty}^{\infty} S_E(k) (1 - \exp(-k^2 \delta^2 \alpha^2)) dk \quad (4.12)$$

A single "universal" curve for $\delta(\tilde{t})$ is not obtainable from this non-linear integro-differential equation. However, solutions can be found as a function of the single parameter $\alpha = \overline{u^2} t_L$.

In Fig. 9 is shown solutions to Eq. (4.12) where $\overline{u^2}$ and $S_E(k)$ corresponds to Fig. 8 and for various values of the integral scale t_L . For all cases shown, the initial puff size $\sigma(0)$ was taken to be 1 metre.

For travel times t that are smaller than, say, 30 sec, the numerical solution of $\sigma(t)$ follows the near field limit of Eq.

(3.69i) and for "intermediate" times when $t_L > 10^5$ s, the spread $\sigma(t)$ continues to follow the prediction in Eq. (4.2) ($\sigma = 0.0123 t^{3/2}$) as long as up to $t \approx 10^4$ s. Still for high values of t_L , the cloud then enters the exponential growth regime and first when $t \gg t_L$ and $\sigma > 10^4$ km the far field limit ($\sigma^2 \propto t$) is ultimately reached.

Values of t_L smaller than $\sim 10^5$ s significantly alter the general behaviour of the growth with time as shown. For t_L as small as ~ 100 s, even not the "intermediate" $3/2$ -region exists. In the literature values of t_L range from 500 to $2 \cdot 10^5$ s (Gifford, 1982). A simple, but very crude estimate based on Pasquill's β -method is: $t_L \approx \beta l / (\overline{u^2})^{1/2}$. Taking $\beta = 4$ and l and $\overline{u^2}$ from Fig. 8, $t_L = 4 \cdot 340 \cdot 10^3 / 30 = 45 \cdot 10^3$ s. When comparison is made with the empirical curve in Fig. 9 of horizontal atmospheric diffusion data, taken from Hage et al. (1967), this value of t_L seems rather high. A time scale of the order ~ 1 hour (3600 s) fits better to the empirical data. At small wavenumbers, the spectral values in Fig. 8, and thereby also the energy $\overline{u^2}$ and the length scale l of the hypothetical spectrum, are maybe unrealistically high, and smaller values hereof would result in better agreement with the empirical curve, when t_L is calculated by the Pasquill β -method.

Before any final conclusion on the relative diffusion theory is drawn from the study here, however, it should be emphasized that

the model Eq. (4.12) used for the computation of $\sigma(t)$ in Fig. 9 is a rather simplified version of the more general theory, Eq. (3.36). In summary, the simplifications involved here are that the two-particle covariance function $R_{abs}(\xi, t)$ has been written as a product $\overline{u^2} \rho_L(\tau) \rho_E(\xi)$. The integral, Eq. (3.59), of $\rho_L(\tau)$ has then been modelled by Eq. (4.10), whereas $\rho_E(\xi)$ is specified through the inverse Fourier transform of $S_E(k)$ in Fig. 8.

Figure 9 finally shows the single-particle diffusion coefficient, $(\overline{u^2})^{1/2} \cdot t$, corresponding to the case where $t_L = \infty$ and an infinite averaging time. As discussed for instance by Mikkelsen and Troen (1981), this coefficient represents an upper limit for σ in the far field limit. This condition is in Fig. 9 seen to be fulfilled whereas the corresponding value in the relative diffusion study by Sheih (1980) was exceeded by a factor of 3.

5. DISCUSSION

Derivation of analytical solutions for the turbulent spreading of a cloud, in terms of the two-particle covariance function (Eq. (3.26)), or in terms of its corresponding spectrum (Eq. (3.47)), were made possible by assuming a non-fluctuating Gaussian particle distribution function. Inclusion of concentration fluctuations C' in the analysis, so that $C = \bar{C} + C'$ and $\overline{C'^2} > 0$ would inevitably have introduced terms in the analysis of the form (in Eq. (3.11) and onward)

$$\overline{u(x_i, t)u(x_j, t-\tau)c'(x_i, t)c'(x_j, t-\tau)} \quad (5.1)$$

together with third order covariances of the variates u and c' as well. Therefore, it is expected that the number as well as the quality of the assumptions required by conventionally modelling such terms (using eddy diffusivities) probably would have introduced at least as much uncertainty, if not even more, as is introduced here by setting $C' = 0$.

There does not seem to exist much reported observation of the mean square of the fluctuations in concentration $\overline{C'^2}$ in clouds, but experimental evidence for steady plumes (summarized on pp. 236-242 of Csanady (1973)) suggests that the distribution of $\overline{C'^2}$ is self-similar and that their ratio to the square of the mean concentration $\overline{C'^2}/\bar{C}^2$ has a value at the centre which varies significantly from experiment to experiment (but typically some-

what less than 0.5) and then increases outwards, reaching values of order ~ 10 at the outer edge of the instantaneous plume.

Chatwin and Sullivan (1979) considered the mean square of the fluctuation in concentration $\overline{C'^2}$ and the ratio $\overline{C'^2}/\overline{C^2}$. The main theme of their paper is the way in which \overline{C} , $\overline{C'^2}$ and $\overline{C'^2}/\overline{C^2}$ vary in space and with time. In terms of the fluid velocity vector \underline{v} relative to the moving origin \underline{c} of a cloud, the Eulerian mass balance over a stationary volume elements reads

$$\frac{\partial C}{\partial t} + \nabla \cdot (C \underline{v}) = \kappa \nabla^2 C \quad (5.2)$$

Here $\nabla \cdot$ and ∇^2 are the divergence and the Lapacian operators in the moving frame, respectively, and κ is the molecular diffusivity. The instantaneous concentration of the cloud can be written in terms of its ensemble means on fluctuations as follows

$$C(y,t) = \overline{C}(y,t) + C'(y,t), \quad \overline{C'} = 0 \quad (5.3)$$

For the relative velocity in the moving frame, it follows from Eq. (2.19) that $\overline{\underline{v}} = 0$, so

$$\underline{v} = \underline{v}'(y,t), \quad \overline{\underline{v}'} = 0 \quad (5.4)$$

Substitution of Eq. (5.3) and Eq. (5.4) into Eq. (5.2) leads in the normal way to the following equations for C and C' :

$$\overline{\frac{\partial C}{\partial t}} + \nabla \cdot (\underline{v}' \overline{C'}) = \kappa \nabla^2 \overline{C} \quad (5.5)$$

and

$$\frac{\partial C'}{\partial t} + \nabla \cdot (\underline{v}' \overline{C} + \underline{v}' C' - \overline{\underline{v}' C'}) = \kappa \nabla^2 C' \quad (5.6)$$

The equation for C'^2 is obtained from Eq. (5.6) by multiplying by $2C'$, assuming incompressibility ($\nabla \cdot \underline{v} = 0$) and taking the ensemble mean. After rearranging, it becomes

$$\begin{aligned} \overline{\frac{\partial C'^2}{\partial t}} = & - 2 \overline{\underline{v}' C'} \nabla \overline{C} \\ & + \nabla \cdot (\kappa \nabla \overline{C'^2} - \overline{\underline{v}' C'^2}) \\ & - 2 \kappa \overline{(\nabla C')^2} \end{aligned} \quad (5.7)$$

The first term on the right hand side is conventionally described as the production of $\overline{C'^2}$ (by feeding from the distribution of \overline{C} through the mechanism described by the term in Eq. (5.5) involving $\overline{\underline{v}' C'}$). The divergence term in Eq. (5.6) has zero integral over all space and, using conventional language, represents the transfer of $\overline{C'^2}$ from place to place. The last term on the right hand side of Eq. (5.6) constitutes a drain for $\overline{C'^2}$ and can be associated with a dissipation rate of the quantity $\overline{C'^2}$. Resemblance of Eq. (5.6) with the equation for turbulent kinetic energy is evident, only is an advection term ($\overline{\underline{v} \cdot \nabla C'^2}$) missing as

a consequence of that reference is made to the moving coordinate system, in which $\underline{\bar{v}} = 0$.

Immediately after the deployment of, say a Gaussian cloud, the concentration distribution $C(x,t)$ resembles that of the initial distribution $C(x,0)$, and since $\overline{C^2} = \bar{C}^2 + \overline{C'^2}$, the ratio $\overline{C'^2}/\bar{C}^2 \approx 0$ in this limit for small t . In the limit for large times, on the other hand, Chatwin and Sullivan shows that, as a consequence almost entirely of molecular diffusion (present through the dissipation rate in Eq. (5.6)), the magnitude of \bar{C} and $\overline{C'^2}$ decay to zero in a way which depends on the details of the fine scale structure of the velocity field. This is probably one reason why experimental measurement of diffusion of gases and heat show that $\overline{C'^2}$ remains of the same order as \bar{C}^2 as plume or clouds develop.

Disregarding for a while the molecular diffusivity κ in Eq. (5.5), it is seen that the statistical theory derived in Chapter 3 is inconsistent with the Eulerian fluid description, when C' and thereby $\nabla \cdot (\underline{\bar{v}}' C')$ $\neq 0$. Therefore, the statistical theory leading to Eq. (3.36) becomes consistent with the fluid description only, when a time-dependent eddy diffusivity $1/2 \, d\sigma^2/dt$ is used to model the flux term $\underline{\bar{v}}' C' = 1/2 \, d\sigma^2/dt \, \nabla C$.

In order to experimentally verify the derived formula for relative diffusion (Eq. (3.36)), the two-particle covariance function $R_{abg}(\xi, \tau)$, or its corresponding spectrum function $S(k, \omega)$, has to be estimated from the turbulent field in question. This

is especially so when the travel time t is of the same order of magnitude as the integral time scale t_L . From a practical point of view, however, this is rather inconvenient, because reliable Lagrangian statistics of a flow-field are difficult, if not impossible to obtain. Hay and Pasquill (1959) proposed a working approximation to circumvent this difficulty by assuming that the Eulerian and Lagrangian auto-covariance functions are similar in shape, and that the ratio of the Lagrangian to the Eulerian time scale β is the only parameter to be determined.

Setting $\xi = 0$, this simple hypothesis may be written in the present notation as

$$R_{abs}(0, \beta\tau) = \tilde{R}_{abs}(0, \tau) \quad (5.8)$$

where \tilde{R}_{abs} refers to an Eulerian (fixed point) auto-covariance function.

It will be proposed here that this simple hypothesis applies to the more general situation as well, where the displacement ξ is different from zero, i.e.

$$R_{abs}(\xi, \beta\tau) = \tilde{R}_{abs}(\xi, \tau)$$

As also argued by Hay and Pasquill (1959), the assumption on precise similarity between the Lagrangian and Eulerian auto-covariance functions is unlikely to produce substantial errors as long as the similarity in shapes are roughly satisfied.

The relation between the spectrum function $S(k, \omega)$ and its corresponding, entirely Eulerian spectrum function, $\tilde{S}(k, \omega)$ is simply obtained by substitution of $\tau = \beta t$ in Eq. (3.40). Thereby

$$S(k, \omega) = \beta \tilde{S}(k, \beta \omega) \quad (5.10)$$

This shows that the shape of the spectrum function $S(k, \omega)$ and the entirely Eulerian spectrum function $\tilde{S}(k, \omega)$ also is found to be similar. In close analogy with Hay and Pasquill's working approximation, Eq. (5.10) implies that the value of the spectrum function S , at a fixed value of k and at the frequency ω , is equal to the Eulerian spectrum function \tilde{S} at wavenumber k , and at frequency $\beta \omega$.

An alternative to direct measurements of the covariance function $R_{abs}(\xi, \tau)$, namely Taylor's suggestion Eq. (1.2), has already been analysed in Section 3.3.

In their study, Smith and Hay (1961) consider the growth of a Gaussian cloud in a three-dimensional, isotropic field of turbulence. However, the covariance function $R_{abs}(\xi, \tau)$ here appears as an entire Eulerian covariance function $\rho_E(\xi + u\tau)$ as a consequence of the following simplifying assumptions: 1) The cloud is assumed to expand "Quasi-stationary", whereby the relative velocity covariance function $R_{rel}(t, \tau) \approx R_{rel}(\tau)$, being a function of the time lag, τ , only. 2) The Lagrangian and the Eulerian covariance functions, $R_{rel}(\tau)$ and $\tilde{R}_{rel}(\tau)$, respectively, are assumed to be similar in shape, the ratio of the respective time scales being β .

In a recent study, Van Buijtenen (1982) proposes two methods to express the mixed space-time covariance function $R_{abs}(\xi, \tau)$ as a function of an Eulerian space covariance function and a time covariance function: one is based on a statistical consideration and one on basis of physical analogy with mixed longitudinal and lateral space correlations. The statistical approach seems to be the more general and useful; the second formula, however, is simpler and can be useful in specific cases.

The above-mentioned methods, all designed to circumvent the difficulty associated with a direct measurement of $R_{abs}(\xi, \tau)$ from the turbulent field in question, seems though to have in common that they suffer from experimental verification.

6. CONCLUSIONS

By assuming Gaussian particle distribution functions, a statistical theory for the turbulent spread of a one-dimensional cloud in homogeneous and stationary have been proposed in terms of a two-particle covariance function $R_{abs}(\xi, \tau)$, cf. Eq. (3.36). A simple working approximation, Eq. (5.9) is suggested for the determination of this covariance function in terms of entirely Eulerian fields.

Applicable for diffusion times that are small compared to the integral time scale of the turbulence, simple expressions for the growth of the puff's standard deviation $\sigma(t)$ have been derived by assuming that the wavenumber spectrum, corresponding to the Eulerian space covariance is a power law δk^p .

For the inertial subrange in atmospheric turbulence, where $p = -5/3$, the predictions (Eq. (3.69) of the cloud growth is found to be consistent with Batchelor's (1950) similarity theory, both at "small" and at "intermediate" times. In addition to the result of similarity theory, also the constant of proportionality between σ^2 and ϵt^3 have been calculated to 0.534, see Eq. (4.2).

For the case of inertial range two-dimensional turbulence, where $p = -3$, the theory predicts exponential growth in agreement with dimensional analysis by Lin (1972).

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APPENDIX A

Important notation

<u> </u>	ensemble average over all realizations of turbulent field
< >	average over particles in cluster = $1/N \sum_{i=1}^N$
'	deviation from cluster mean
<u>a</u>	vector quantity of magnitude a
c	centre of mass position of the cloud, and origin for the moving frame co- ordinate, y
C	concentration distribution function of cloud
d	linear extension of a fluid particle
G	Gaussian particle distribution function

H	Heavyside's step function
i	(as suffix) designation of individual particles (x_i) or fluid element (dx_i) in the cloud
k	wavenumber ($= 2\pi/\lambda$ where λ is wavelength)
k_r	eddy diffusivity for relative diffusion
K	eddy diffusivity for absolute diffusion
l	distance separating two typical marked fluid elements
l_E	Eulerian integral length scale
M	number of fluid particles in the turbulent field
N	number of tracer particles in a cloud
n	difference in particle number i-j

p	exponent in power function k^p
$P(x', x'', t)$	two-particle displacement probability density, see Eq. (2.14)
q	distance neighbour function, see Eq. (1.2)
Q	total amount of matter released with a cloud
r	relative velocity correlation function, see Eq. (2.22)
$R_{abs}(\tau)$	Lagrangian auto-covariance function, $\overline{u(t)u(t-\tau)}$
$R_{abs}'(\xi, \tau)$	two-particle velocity covariance function- see Eq. (3.36). (Note that $R_{abs}(0, \tau) \equiv R_{abs}(\tau)$)
$R_{cm}(t, \tau)$	centre of mass covariance function, see Eq. (3.6)
$R_{rel}(t, \tau)$	relative velocity covariance function, see Eq. (3.6) (Note $R_{rel}(t, \tau) = t_r \cdot \overline{v^2(t)}$)
$S(k, \omega)$	spectrum function corresponding to covariance $R_{abs}(\xi, \tau)$

$S_E(k)$	spectrum function corresponding to $\rho_E(\xi)$
$S_L(\omega)$	spectrum function corresponding to $\rho_L(\tau)$
t	time, with origin at moment of release of cloud
t_E	fixed frame (Eulerian) integral time scale
t_L	fixed frame (Lagrangian) integral time scale
$t_r(t)$	Lagrangian integral time scale appropriate for relative diffusion, see Eq. (2.23)
T_c	time scale $\eta^{-1/3}$ in the enstrophy inertial subrange
u	velocity component referred to fixed frame (x)
v	velocity component referred to moving frame (y)

V_{cm}	centre of mass velocity component dc/dt
x	fixed or absolute frame coordinate
y	moving or relative frame coordinate
α	$(\overline{u^2}t_L)^{1/2}$, see Eq. (4.11)
β	ratio between Lagrangian and Eulerian integral time scales, viz. t_L/t_E
δ	coefficient to power law $S_E(k) = \delta k^p$ with dimension $m(1+p)$
$\Gamma(p)$	gamma function $\int_0^\infty x^{p-1} e^{-x} dx$
ϵ	rate of dissipation of energy
η	enstrophy cascade rate
κ	molecular diffusivity
μ	fourth moment of the concentration distribution about the centre of mass
ν	kinematic viscosity

$\xi_{ij}(t)$	separation of two particles i, j at fixed time, see Eq. (3.22)
$\rho_E(\xi)$	Eulerian correlation coefficient $R_{abs}(\xi, 0)/\overline{u^2}$
$\rho_L(\tau)$	Lagrangian correlation coefficient $R_{abs}(0, \tau)/\overline{u^2}$
σ	standard deviation of the particle positions about the centre of the Gaussian puffs
σ_0	initial puff size $\sigma(t = 0)$
τ	time lag, see Fig. 1
ω	angular frequency ($= 2\pi \times$ cycles per unit time)

APPENDIX B

Spectral definitions

This appendix justifies some of the spectral relations used in the body of the report.

The spectrum of the two-particle, mixed Lagrangian-Eulerian covariance function $R_{abs}(\xi, \tau)$ is defined through the Fourier transform pair

$$S(k, \omega) = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \exp(-i(k\xi + \omega\tau)) d\xi d\tau \quad (B.1)$$

$$R_{abs}(\xi, \tau) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i(k\xi + \omega\tau)) dk d\omega \quad (B.2)$$

By definition, $R_{abs}(0,0) = \overline{u^2}$, so from Eq. (B.2) with $(\xi, \tau) = (0,0)$ we have

$$\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) dk d\omega = \overline{u^2} \quad (B.3)$$

For the case where $\xi = 0$, which corresponds to an entirely Lagrangian (single particle) correlation function $\rho_L(\tau) = \overline{u(x,t) u(x,t-\tau)} / \overline{u^2}$, we also define the Fourier transform pair

$$S_L(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \rho_L(\tau) \exp(-i\omega\tau) d\tau \quad (B.4)$$

$$\rho_L(\tau) = \int_{-\infty}^{\infty} S_L(\omega) \exp(i\omega\tau) d\omega \quad (B.5)$$

Since $\rho_L(0) = 1$, we have from Eq. (B.5)

$$\int_{-\infty}^{\infty} S_L(\omega) d\omega = 1$$

Analogously, for the case where $\tau = 0$, which corresponds to an entirely Eulerian two-point correlation function $\rho_E(\xi) = \overline{u(x,t) u(x+\xi,t)} / \overline{u^2}$ we define the Fourier transform pair

$$S_E(k) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \rho_E(\xi) \exp(-ik\xi) d\xi \quad (B.6)$$

$$\rho_E(\xi) = \int_{-\infty}^{\infty} S_E(k) \exp(ik\xi) dk \quad (B.7)$$

Since $\rho_E(0) = 1$, we have from Eq. (B.7)

$$\int_{-\infty}^{\infty} S_E(k) dk = 1 \quad (B.8)$$

By noting that $R_{abs}(0, \tau) = \rho_L(\tau) \cdot \overline{u^2}$, we get from Eq. (B.2), by setting $\xi = 0$

$$\overline{u^2} \rho_L(\tau) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i\omega\tau) dk d\omega \quad (B.9)$$

A comparison with Eq. (B.4) now shows the relation

$$\overline{u^2} S_L(\omega) = \int_{-\infty}^{\infty} S(k, \omega) dk \quad (B.10)$$

Analogously, by noting that $R_{abs}(\xi, 0) = \rho_E(\xi) \cdot \overline{u^2}$, we get from Eq. (B.2) with $\tau = 0$, and Eq. (B.7) the relation

$$\overline{u^2} S_E(k) = \int_{-\infty}^{\infty} S(k, \omega) d\omega$$

FIGURE LEGENDS

Fig. 1. Qualitative behaviour of relative velocity correlation $r(\tau, t)$ for given t . The shaded area visualizes the relative integral time scale $t_r(t)$.

Fig. 2. The motion of a Gaussian cloud G in the fixed frame of reference x as a function of time t . The centre of mass coordinate of the cloud c defines the origin of the moving frame y , relative to which the clouds dispersion in terms of the standard deviation σ is defined. Also shown are the two fixed points in the moving frame, y' and y'' , on which the covariance function $\overline{u(y'+c, t)u(y''+c, t-\tau)}$ depends.

Fig. 3. The moving frame trajectory y_i of a marked fluid particle (i) that at time $t-\tau$ holds the position $y_i(t-\tau)$. The quantity $\Delta y_i \equiv y_i(t) - y_i(t-\tau)$ as well as its distribution function $G_{\Delta y_i}$ is shown at time t .

Fig. 4. The trajectory of an arbitrary fluid particle (j), which at time $t-\tau$ is in the position $y_j(t-\tau)$ and another particle (i), which at the same time holds a position displayed the distance ξ_{ij} relative to (j). Note that ξ_{ij} denotes the separation of the two particles in both the moving and the fixed frames: $\xi_{ij} = y_i(t-\tau) - y_j(t-\tau) = x_i(t-\tau) - x_j(t-\tau)$. Otherwise as in Fig. 3.

Fig. 5. The two-particle covariance function defined in Eq. (3.26). a) Referring the fixed particle separation ξ_{ij} to time $t-\tau$: $\overline{u(x_i(t))u(x_i(t-\tau)-\xi_{ij}(t-\tau))}$. b) Referring ξ_{ij} to time t : $\overline{u(x_j(t-\tau))u(x_j(t)+\xi_{ij}(t))}$. In homogeneous and stationar turbulence, these two definitions are identical.

Fig. 6. Iso-contour plot of a hypothetical spectrum $S(k, \omega)$. Its maximum value is at $(k, \omega) = (0, 0)$ from where the function monotonically decreases through the levels I, II and III. The cut-off frequency associated with the low-pass filter $\sin(\omega t)/\omega t$ is schematically drawn as the vertical line at $\omega = t^{-1}$. Correspondingly, the high-pass filter $(1 - \exp(-k^2 \sigma^2))$ essentially cuts away wavenumbers that are smaller than σ^{-1} . The shaded area therefore represents the part of the spectrum $S(k, \omega)$ that essentially contributes to the integral over k and ω in Eq. (3.44).

Fig. 7. The four regimes divided by the length scale ℓ and time scale t_L in a time-space plot of relative diffusion, based on a power law spectrum $S_E(k)$ that is proportional to k^{-3} for $k > 1/\ell$, and of constant amplitude for $k < 1/\ell$.

Fig. 8. Summation of relative diffusion $\sigma(t)$ of an initially small Gaussian cloud in relation to a schematic wavenumber spectrum composed from the literature by K.S. Gage, 1979, D.K. Lilly and E.L. Petersen, 1983, and D.K. Lilly, 1983. See text for description.

Fig. 9. Plot of cloud size, σ , for various values of the Lagrangian integral time scale t_L vs. travel time t , according to Eq. (4.12) and the energy spectrum of Fig. 8. The dotted curve, see Hage et al. (1967), illustrates an empirical curve of horizontal atmospheric diffusion data over the entire atmospheric range. The maximum single particle diffusion coefficient, $(\overline{u^2})^{1/2}t$, corresponding to the case where $t_L = \infty$, is shown as the topmost dashed-dotted line (see also Mikkelsen and Troen (1981)).

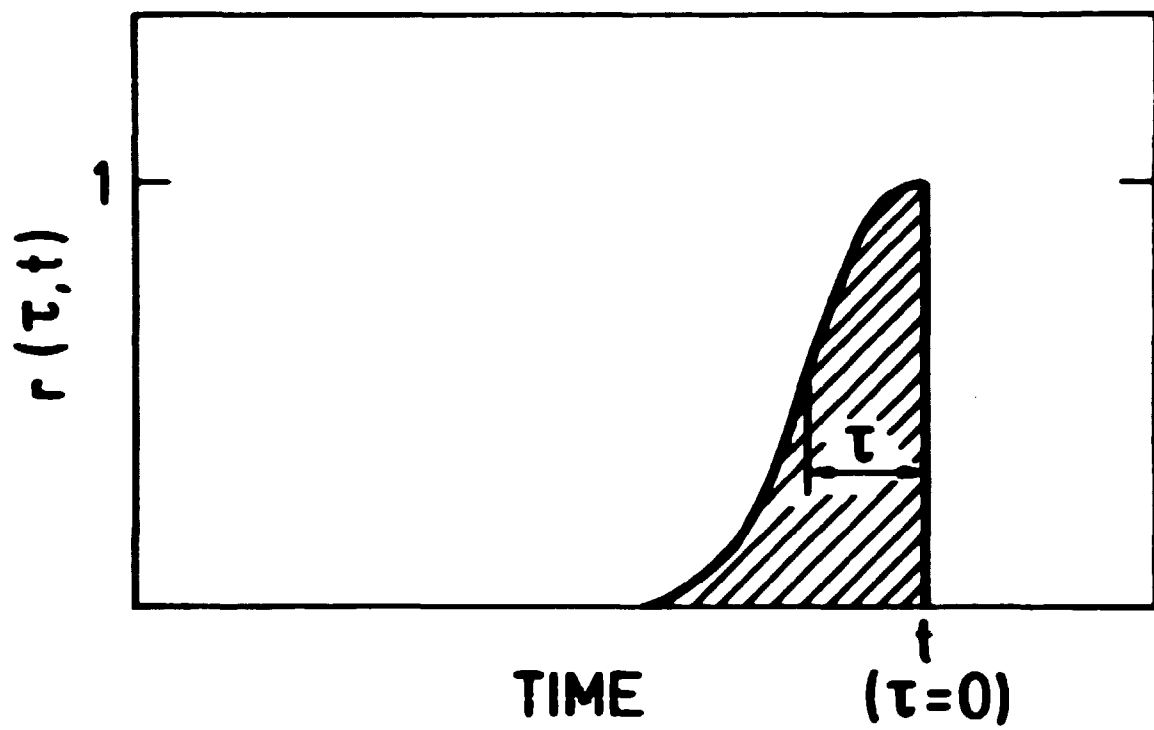


Fig. 1

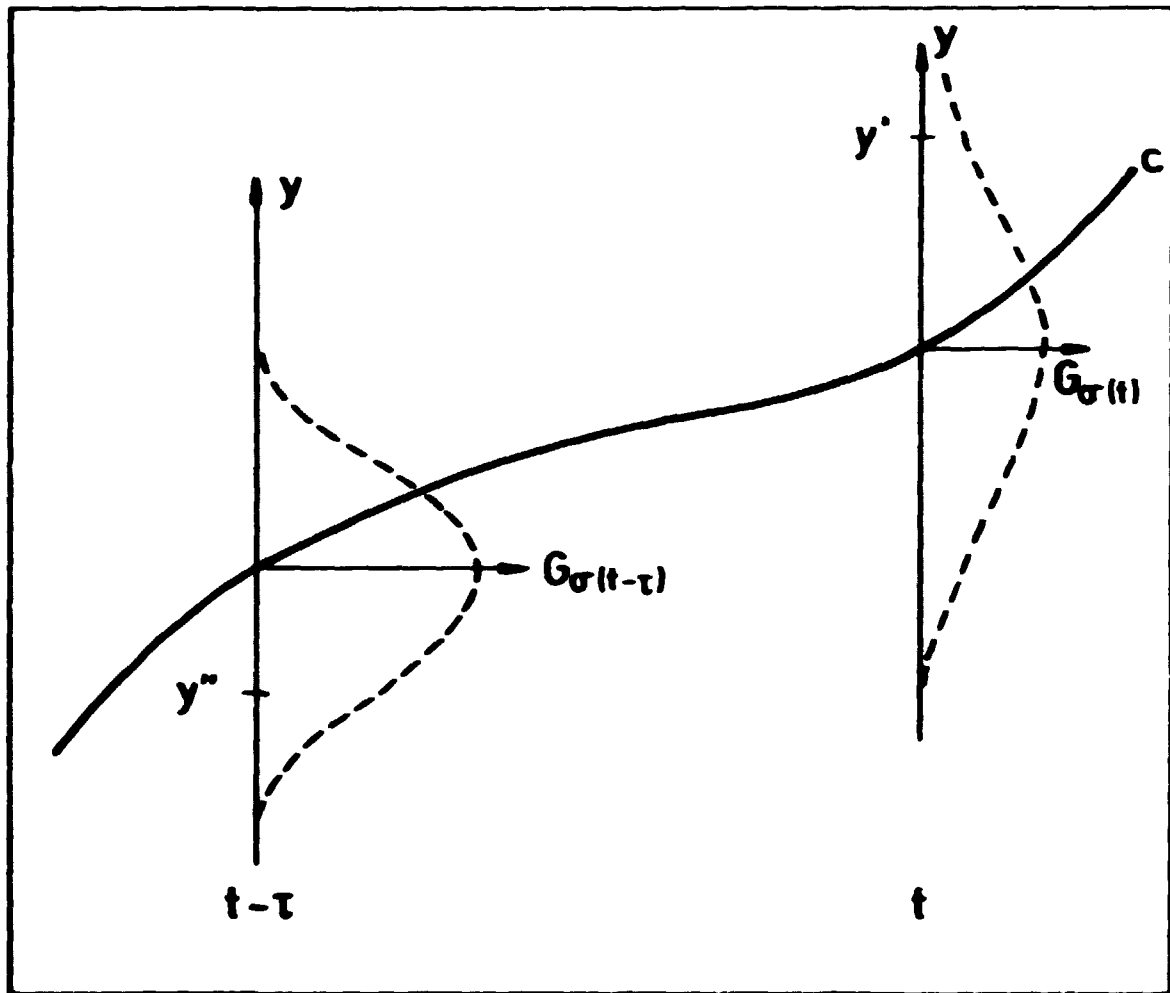


Fig. 2

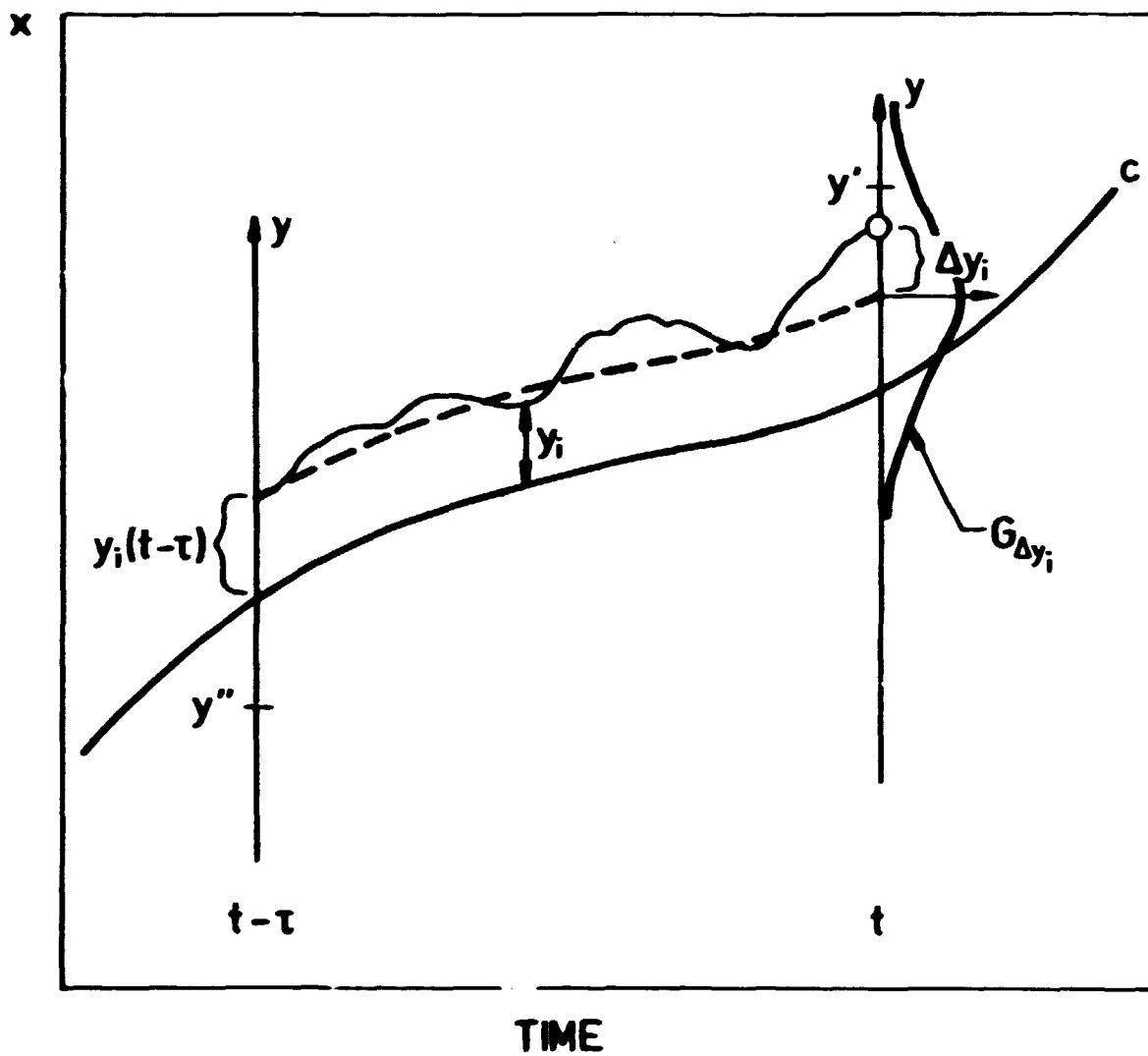


Fig. 3

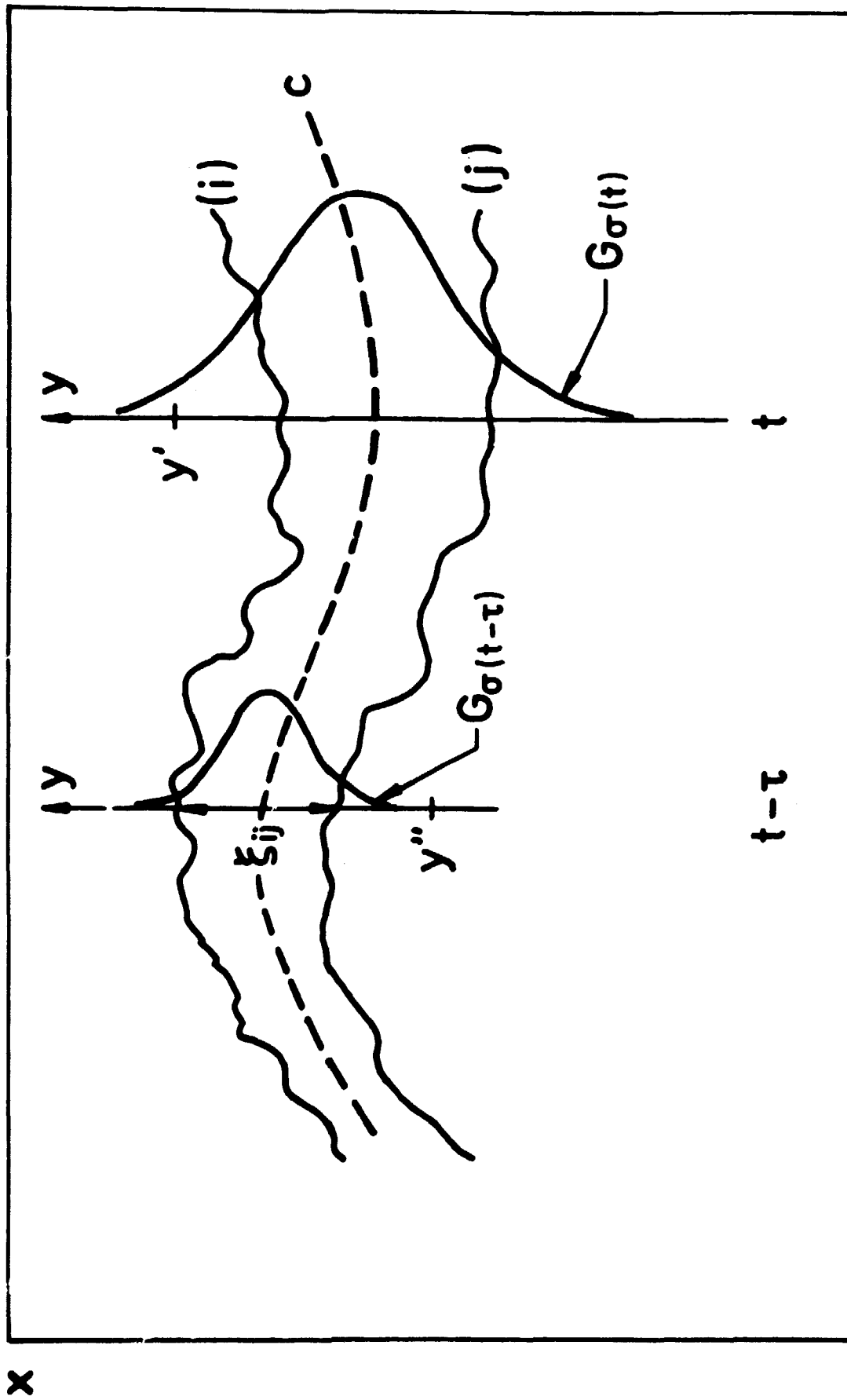


Fig. 4

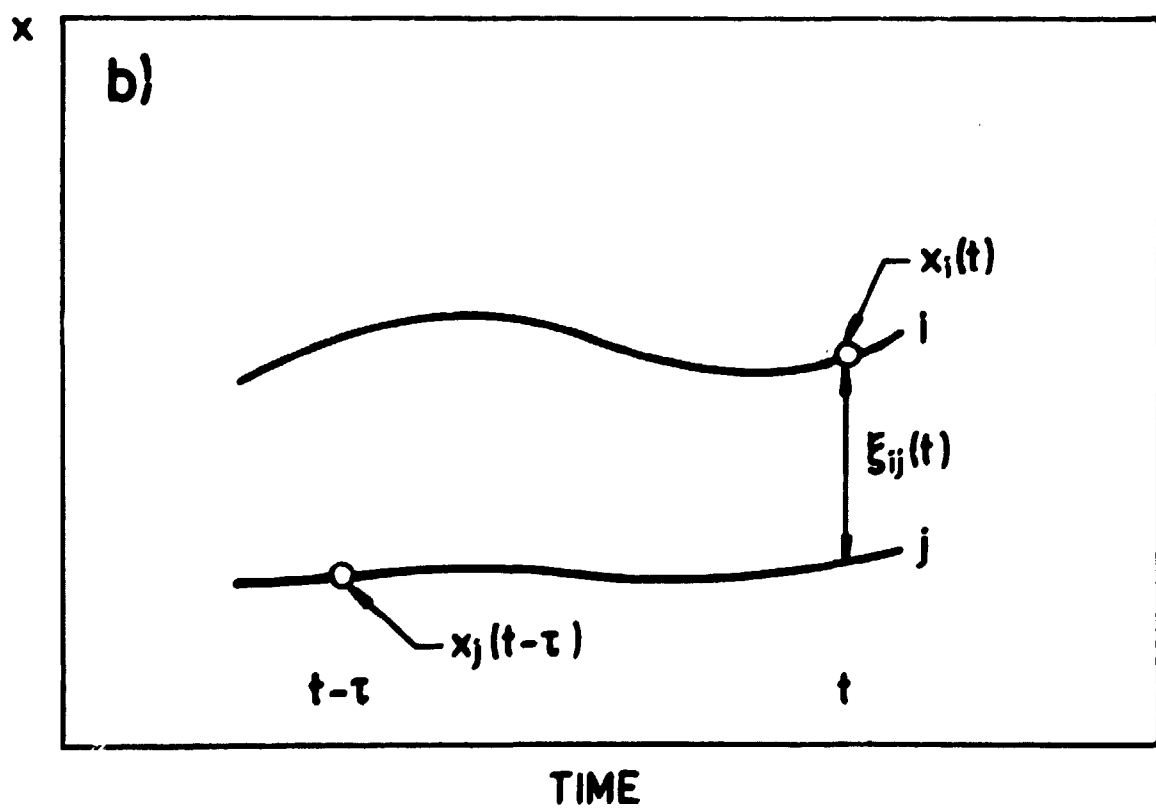
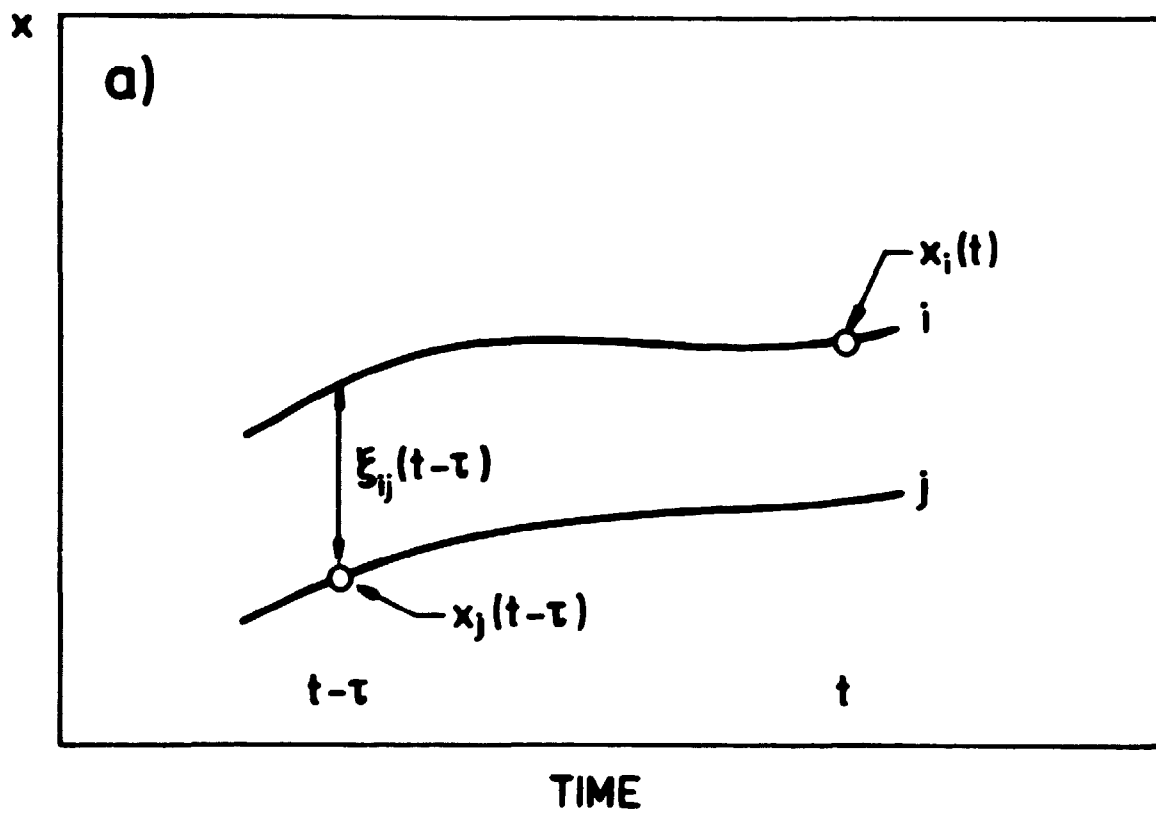


Fig.5

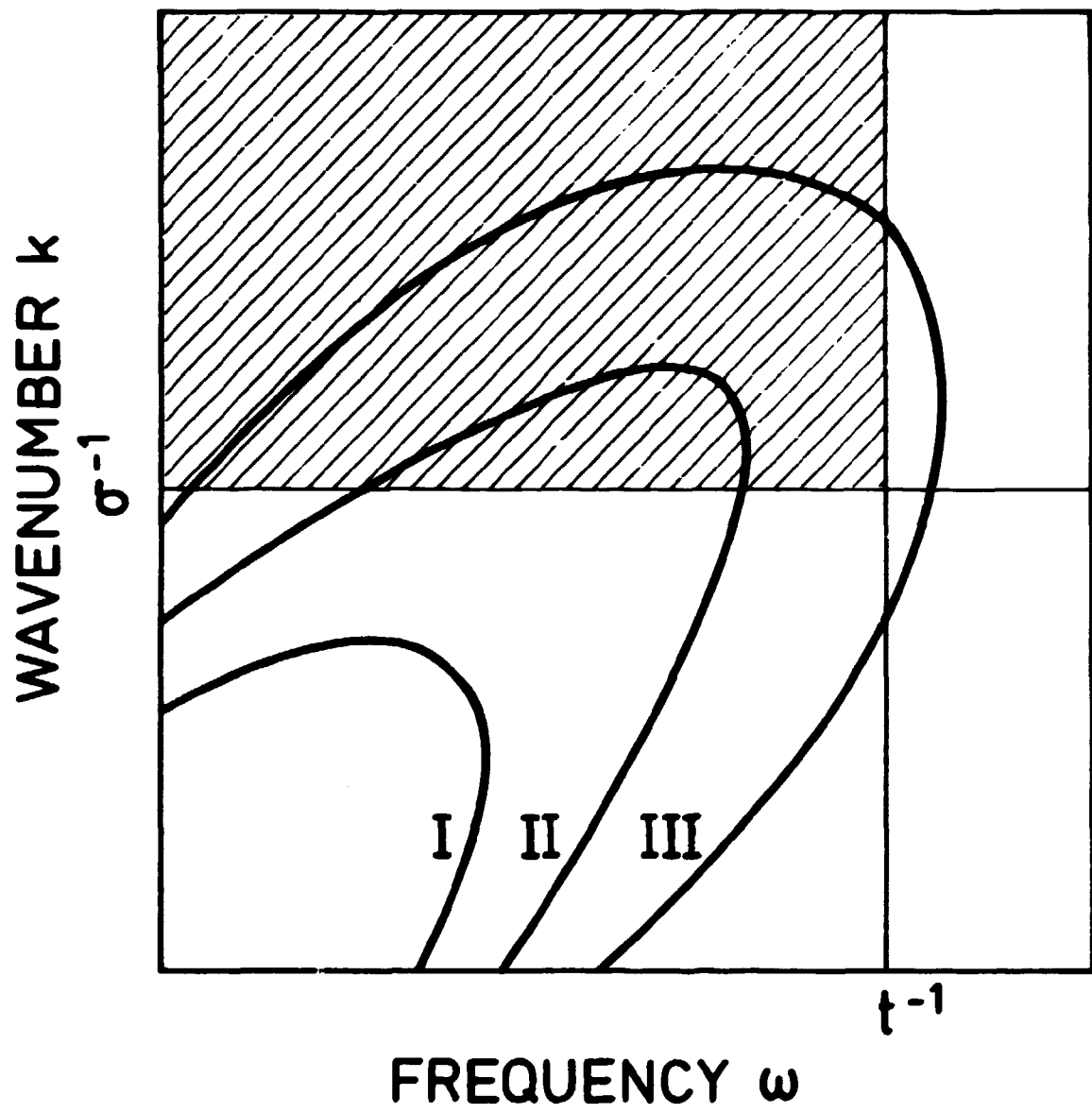


Fig. 6

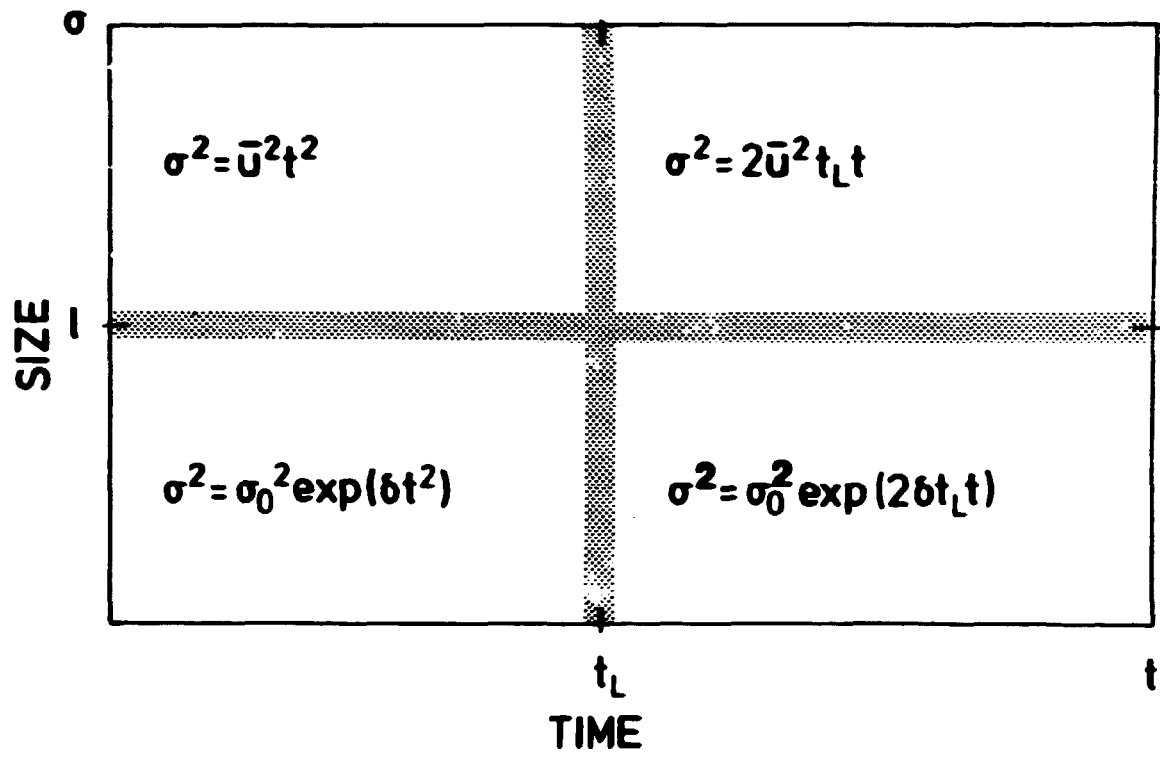


Fig. 7

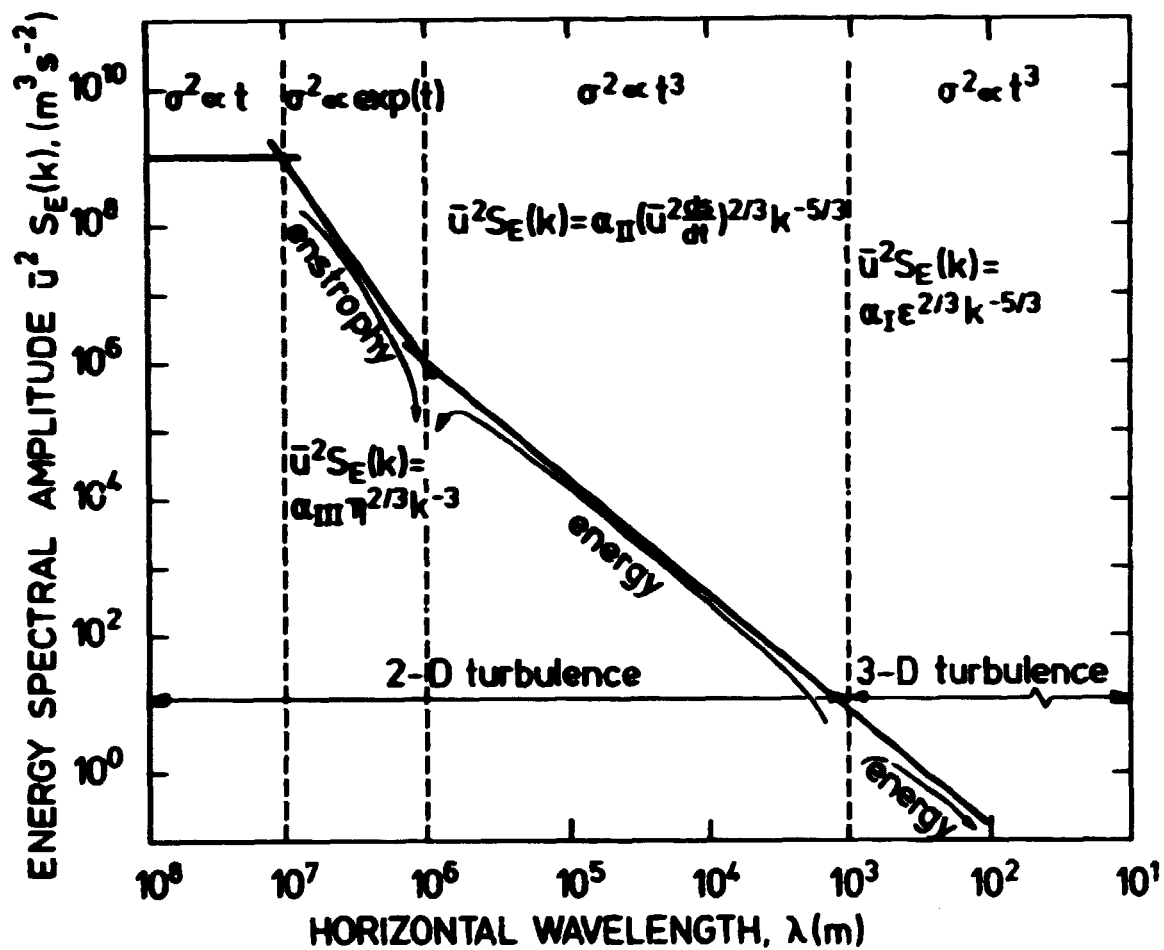


Fig. 8

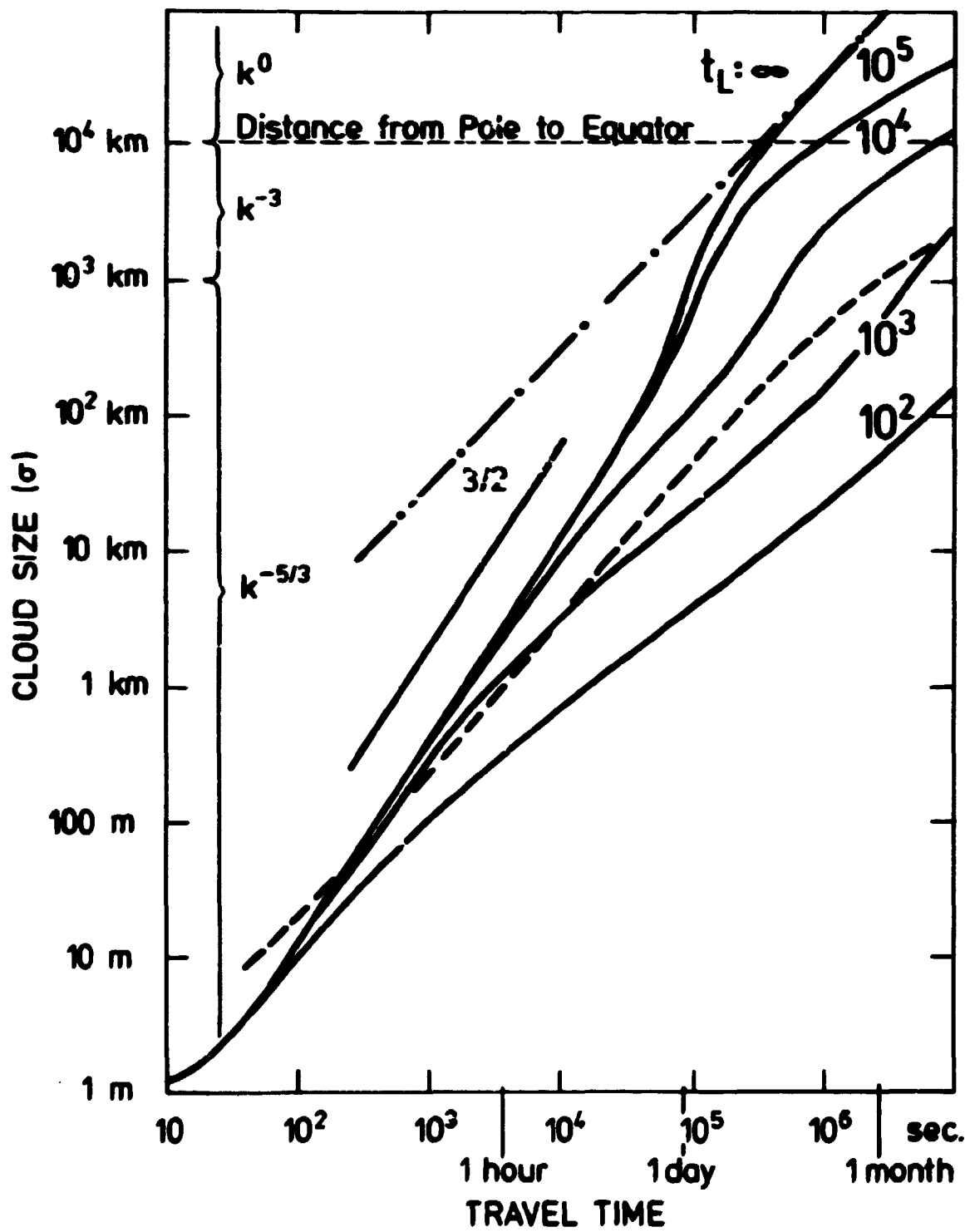


Fig. 9

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Title and author(s)

A Statistical Theory on the Turbulent
Diffusion of Gaussian Puffs

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Date December 1982

Department or group

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Group's own registration
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Abstract

The relative diffusion of a one-dimensional Gaussian cloud of particles is related to a two-particle covariance function $R_{abs}(\xi_{ij}, \tau) = u(x_i(t)u(x_i(t-\tau) - \xi_{ij}))$ in a homogenous and stationary field of turbulence. This two-particle covariance function expresses the velocity correlation between one particle (i) which at time t is in the position x_i , and another particle (j), which at the previous time t- τ is displaced the fixed distance ξ_{ij} relative to $x_i(t-\tau)$. For $\xi_{ij} = 0$, R_{abs} reduces to the Lagrangian covariance function of a single particle. Setting, on the other hand, the time lag τ equal to zero, a pure Eulerian fixed point covariance function results.

For diffusion times that are small compared to the integral time scale of the turbulence, simple expressions are derived for the growth of the clouds standard deviation $\sigma(t)$ by assuming that the wave number spectrum corresponding to the Eulerian space covariance $R_{abs}(\xi_{ij}, 0)$ can be expressed as a power law δk^p , where δ is a constant. For instance, by setting $p = -5/3$, an initially small cloud is found to grow as $\sigma^2(t) = (2\Gamma(\frac{2}{3})\delta)^{3/2} t^3$ in agreement with

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Batchelor's (1950) inertial subrange theory. Correspondingly, for the enstrophy cascade subrange in two-dimensional turbulence, for which case $p = -3$, the theory yields $\sigma^2(t) = \sigma_0^2 \exp(\epsilon t^2)$, where σ_0 denotes the initial size of the cloud.